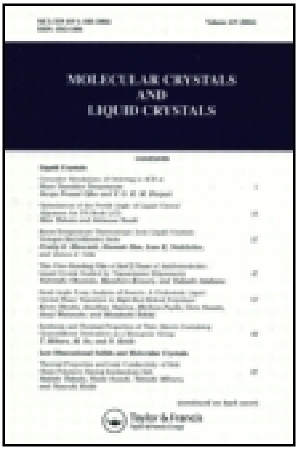

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Analytical Solutions for Small Distortion Waves in the Freedericks' Transition Problem

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Analytical Solutions for Small Distortion Waves in the Freedericks' Transition Problem†

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Analytical general solutions for small distortion waves in the Freedericks' transition problem have been obtained for steady state flow. Inconsistencies in solutions found in the literature are removed under the assumption that the linear and angular momenta of an incompressible fluid are conserved within the hydrodynamic limit. The time dependence of the director of the nematic phase and its distortion pattern in the presence of a magnetic field is thus obtained.

I. INTRODUCTION

In an unperturbed non-chiral liquid crystal state, the molecules are aligned in a preferred direction. In the presence of a magnetic or electric field, the system will undergo a Freedericks' transition in which the molecules assume a distorted pattern. Planar motion of the molecules occurs as the sample reorients itself. In the words of hydrodynamic and curvature elastic theories, distortion waves of the director are coupled with the hydrodynamic velocities in the system.

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Much work has been done in this field, both theoretically and experimentally. The most representative work has been done by Berreman,[1-4] the Orsay group (Pieranski, Brochard, Guyon),[5-8] Van Dorn,[9] Schadt,[10] Helfrich,[11] Penz and Ford,[12] Ben-Abraham,[13] Leslie,[14] and others. In all instances, studies were made for the cases in which different boundary conditions were imposed under either magnetic or electric field or for relative motion between the contacting plates of the thin film. The fluid instability of twisted cells and bend and splay modes of distortion waves were solved in both static and dynamic cases.

Analytical solutions for dynamic cases were only obtained in small distortion approximations. The problems arose when examining the physics and equilibrium states of the system, because they overlooked the mathematical consequences of their assumptions. They assumed that linear and angular momenta of an incompressible fluid are conserved within the hydrodynamic limit. Due to the inconsistencies which occurred in this approach, a more detailed examination of the physical principles, along with the mathematics of the distortion wave and backflow in the Freedericks' transition problem, is necessary.

Recently, attention in this field has focused on lyotropic systems.[15-17] In particular, laser light scattering studies of magnetic birefringence and mechanical properties of amphiphilic liquid crystal systems are being done by Saupe *et al.*[18] In order to formulate a complete theory and to perform a computer modeling scheme for our problem, we have to give more general and consistent solutions to the distortion waves of the director. Once the basic equations are solved, we will use them as a starting point for our future investigations into large distortion waves, optical and mechanical properties of those lyotropic systems in which we are interested. In this work we present the general solutions to the time-dependent and distortion profiles of the director and the hydrodynamic velocities as the first in a series of three papers with the other two to follow.[19,20]

II. BASIC EQUATIONS AND LINEARIZED SOLUTIONS

To start with, we consider a thin film (thickness ~0.1 mm) incompressible nematic system in the presence of a static magnetic field H, with magnitude equal to, or slightly greater than, the critical field H_c, which gives rise to the Freedericks' transition. (In general, we use a time-dependent field, but for simplicity we start with a purely static field.) The director **L** makes an angle θ with the z-axis and lies in the z-x plane as shown in Figure 1. The

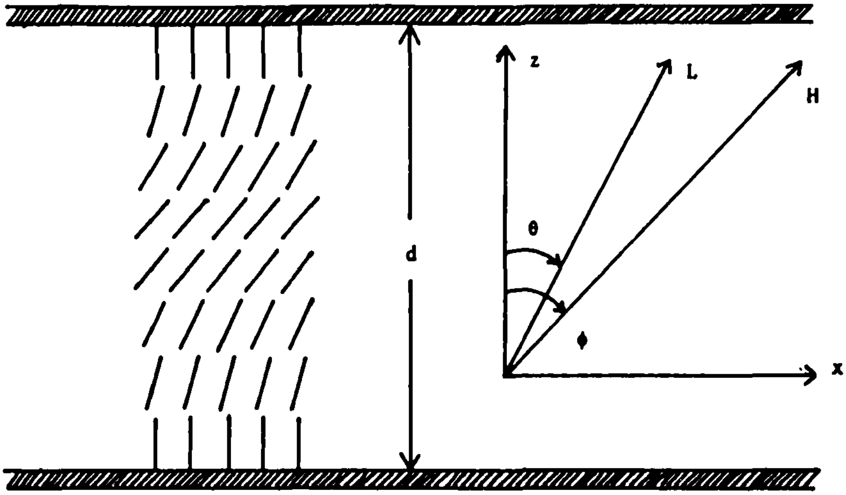

FIGURE 1 Distortion pattern of a homeotropic case.

angle θ is a function of z and time t. The field makes an angle ϕ with the z-axis and also lies in the z-x plane. The field H has an x-component in order to stabilize the director orientations to obtain a uniform (in coordinates of x and z only) distortion pattern. The molecules are fixed normally to the surface and the hydrodynamic velocity has only an x-component. Thus we have the following boundary conditions at the surface:

$$\mathbf{L} = (0, 0, 1)$$
$$\mathbf{v} = (0, 0, 0) \tag{1}$$

In the film,

$$\mathbf{L} = (\sin\theta, 0, \cos\theta)$$
$$\mathbf{v} = (v_x, 0, 0)$$

Based on the Frank and Oseen curvature elastic theory[21-22] and Erikson's and Leslies' hydrodynamic theory[23-26] of nematic liquid crystals, one can calculate the viscous torque (τ_{vis}), elastic torque (τ_{el}), and magnetic torque (τ_H):

$$\tau_{\text{vis}} = (\alpha_2 - \alpha_3)\mathbf{L} \times \mathbf{N} + (\alpha_5 - \alpha_6)\mathbf{L} \times \mathbf{A} \cdot \mathbf{L} \tag{3}$$

Here $\mathbf{N} \equiv d\mathbf{L}/dt - \boldsymbol{\omega} \times \mathbf{L}$ is the director velocity related to the fluid; $\boldsymbol{\omega} = \frac{1}{2}\nabla \times \mathbf{v}$ is the angular velocity of the director; and $\mathbf{A}$ denotes the strain rate tensor.

$$A_{ik} = \frac{1}{2}\left(\frac{\partial v_i}{\partial x_k} + \frac{\partial v_k}{\partial x_i}\right)$$

and α_i's are the Leslie viscous coefficients.

$$\tau_{el} = -k_{33}\nabla^2\mathbf{L} \times \mathbf{L} + (k_{33} - k_{11})\nabla(\nabla \cdot \mathbf{L}) \times \mathbf{L} + (k_{33} - k_{22})[-2(\mathbf{L} \cdot \nabla \times \mathbf{L})\nabla \times \mathbf{L} + \mathbf{L} \times \nabla(\mathbf{L} \cdot \nabla \times \mathbf{L})] \times \mathbf{L} \quad (4)$$

Here k_{ii}'s are the elastic coefficients corresponding to the splay, twist, and bend modes.

$$\tau_H = \chi_a \mathbf{L} \cdot \mathbf{H}(\mathbf{L} \times \mathbf{H}) \quad (5)$$

Here $\chi_a = \chi_\parallel - \chi_\perp$ is the anisotropic magnetic susceptibility of the system. The balance of these torques (i. e., neglecting the inertia torque term $\partial^2\theta/\partial t^2 = 0$) gives

$$[k_{11} + (k_{33} - k_{11}) \cos^2 \theta]\frac{\partial^2\theta}{\partial z^2} - \frac{1}{2}(k_{33} - k_{11}) \sin 2\theta\left(\frac{\partial\theta}{\partial z}\right)^2 - (\alpha_3 - \alpha_2)\frac{\partial\theta}{\partial t} + \frac{1}{2}[(\alpha_3 - \alpha_2) - (\alpha_2 + \alpha_3) \cos 2\theta]\frac{\partial v}{\partial z} - \frac{1}{2}\chi_a H^2[\sin(2\theta + 2\phi)] = 0 \quad (6)$$

By neglecting the inertial force term (i. e., $\partial v/\partial t = 0$) and using a unit volume element of the nematic fluid, the equation of motion[27–29] becomes

$$\rho\frac{\partial v_i}{\partial t} = f_i + \frac{\partial \sigma_{ji}}{\partial x_j} \quad (7)$$

$$[(\eta_2 - \alpha_3) + \frac{\alpha_1}{8} \sin^2 2\theta]\frac{\partial^2 v}{\partial z^2} + \alpha_1 \sin 2\theta\left(\cos^2 \theta - \frac{1}{2}\right)\frac{\partial\theta}{\partial z}\frac{\partial v}{\partial z} - (\alpha_2 + \alpha_3) \sin 2\theta\frac{\partial\theta}{\partial z}\frac{\partial\theta}{\partial t} + [(\alpha_2 + \alpha_3) \cos^2 \theta - \alpha_2]\frac{\partial^2\theta}{\partial z\, \partial t} = 0 \quad (8)$$

where ρ is the density, f_i the external body force per unit volume, σ_{ji} the stress tensor, and η_i the shear viscosity.

In a small distortion approximation, Eqs. 6 and 8 become linear equations which are given by

$$k_{33}\frac{\partial^2\theta}{\partial z^2} - (\alpha_3 - \alpha_2)\frac{\partial\theta}{\partial t} - \alpha_2\frac{\partial v}{\partial z} + \chi_a H^2\left[(-\cos 2\phi)\theta + \frac{1}{2}\sin 2\phi\right] = 0 \tag{9}$$

$$\frac{\partial}{\partial z}\left[(\eta_2 - \alpha_3)\frac{\partial v}{\partial z} + \alpha_3\frac{\partial\theta}{\partial t}\right] = 0 \tag{10}$$

Now solving Eq. (9) for $\partial v/\partial z$, we have

$$\frac{\partial v}{\partial z} = \frac{k_{33}}{\alpha_2}\frac{\partial^2\theta}{\partial z^2} - \frac{(\alpha_3 - \alpha_2)}{\alpha_2}\frac{\partial\theta}{\partial t} + \frac{\chi_a H^2}{\alpha_2}\left[(-\cos 2\phi)\theta + \frac{1}{2}\sin 2\phi\right] \tag{11}$$

Substituting Eq. 11 into Eq. 10, we have an equation with simplified notation

$$[aD_z^2 - bD_t + c]\phi = 0 \tag{12}$$

where

$$a = \frac{k_{33}}{\alpha_2}(\eta_2 - \alpha_3), \qquad b = \frac{1}{\alpha_2}(\alpha_3 - \alpha_2)(\eta_2 - \alpha_3) - \alpha_3$$

$$c = -\frac{\chi_a H^2}{\alpha_2}(\eta_2 - \alpha_3)\cos 2\phi, \qquad D_t = \frac{\partial}{\partial t} \tag{13}$$

$$D_z = \frac{\partial}{\partial t}, \qquad \phi = D_z\theta$$

We write Eq. 12 in an apparent Lagrange form

$$[D_z - bD_t + c]\phi = [D_z - aD_z^2]\phi \equiv f(z, t), \tag{14}$$

which has the characteristic equations

$$\frac{dz}{1} = \frac{dt}{-b} = \frac{d\phi}{f(z, t) - c\phi} \tag{15}$$

Solving the first equality yields

$$t + bz = u \tag{16}$$

where u is a parameter. Geometrically, from Figure 2, one can see that the plane P intersects the solution surface $\phi(z, t)$, the surface of the defined function $f(z, t)$ and the z-t plane. Clearly, the intersection with the z-t plane

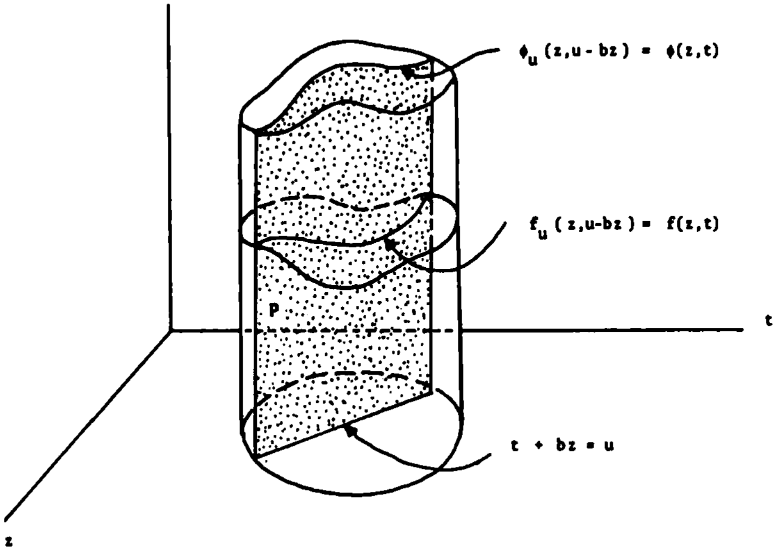

FIGURE 2 Geometrical representation of a characteristic solution.

represented by Eq. 16 determines the character of the intersections of $f(z, t)$ and $\phi(z, t)$. This gives the particular function $f_u(z, u\text{-}bz)$ and the solution $\phi_u(z, u\text{-}bz)$. Guided by this geometrical concept, the general solution of Eq. 12 is found:

$$\theta = \int e^{-cz} g(bz + t)\, dz + \frac{c_1(t)}{x_1} e^{x_1 z} + \frac{c_2(t)}{x_2} e^{x_2 z} + c_3(t) \tag{17}$$

where g is an arbitrary function of $bz + t$, the $c_i(t)$ ($i = 1, 2, 3$) are arbitrary functions of t, and

$$x_{1,2} = \frac{1}{2a}(-1 \pm \sqrt{1 - 4ac}) \tag{18}$$

To solve for v, we need to calculate $\partial\theta/\partial t$ and $\partial^2\theta/\partial z^2$. After substituting these derivatives into Eq. 11, we then integrate it to find v.

$$\alpha_2 v = -ck_{33} \int e^{-cz} g(bz + t)\, dz + bk_{33} \int e^{-cz} g'(bz + t)\, dz$$
$$- (\alpha_3 - \alpha_2) \int\int e^{-cz} g'(bz + t)\, d^2z$$

$$- A \iint e^{-cz} g(bz + t)\, d^2z + k_{33}[c_1(t)e^{x_1 z} + c_2(t)e^{x_2 z}]$$

$$- (\alpha_3 - \alpha_2)\left[\frac{c_1'(t)e^{x_1 z}}{x_1^2} + \frac{c_2'(t)e^{x_2 z}}{x_2^2} + c_3'(t)z\right]$$

$$- A\left[\frac{c_1(t)e^{x_1 z}}{x_1^2} + \frac{c_2(t)e^{x_2 z}}{x_2^2} + c_3(t)z\right]$$

$$+ Bz + C_4(t), \qquad \text{where} \qquad A = \chi_a H^2 \cos 2\phi$$

$$B = \chi_a H^2 \sin 2\phi \tag{19}$$

Equations 17 and 19 are the general solutions subject to the conditions

$$\frac{\partial v}{\partial t} = 0 \text{ for all } z, \qquad v(z = z_0) = 0$$

$$\theta(z = z_0) = 0, \qquad \left.\frac{\partial \theta}{\partial t}\right|_{z=z_0} = 0 \tag{20}$$

The solutions for θ and v both contain terms that involve $g(bz + t)$. Any function $g(bz + t)$ will work subject to the boundary conditions, but we wish to express g in terms of the c_i's. The c_i's will be determined later. In order to do this, we apply the condition $\theta(z_0) = 0$. Because of continuum theory, this can be generalized to $\lim_{\varepsilon \to 0} \theta(z = z_0 + \varepsilon) = 0$, which gives

$$g(z_0) = -k_{33}^{-1} e^{cz_0}[k_{33}(c_1 e^{x_1 z_0} + c_2 e^{x_2 z_0}) + Bz_0 + c_4] \tag{21}$$

Equation 21 represents two conditions since $z_0 = \pm d/2$. A further condition can be found on g by applying conservation of linear momentum $\partial v/\partial t = 0$, which is good for all z; i.e.,

$$0 = -ck_{33} \int e^{-cz} g'(bz + t)\, dz + bk_{33} \int e^{-cz} g''(bz + t)\, dz$$

$$- (\alpha_3 - \alpha_2) \iint e^{-cz} g''(bz + t)\, d^2z$$

$$- A \iint e^{-cz} g'(bz + t)\, d^2z + k_{33}(c_1' e^{x_1 z} + c_2' e^{x_2 z})$$

$$- (\alpha_3 - \alpha_2)\left[\frac{c_1'' e^{x_1 z}}{x_1^2} + \frac{c_2'' e^{x_2 z}}{x_2^2} + c_3'' z\right]$$

$$- A\left[\frac{c_1' e^{x_1 z}}{x_1^2} + \frac{c_2' e^{x_2 z}}{x_2^2} + c_3' z\right] + c_4' \tag{22}$$

Similarly, by applying conservation of angular momentum $\partial^2\theta/\partial t^2 = 0$, another restriction among the g and all c_i's can be obtained.

The second set of solutions are obtained by first integrating Eq. 10 and substituting it into Eq. 9, then solving the resulting equation. The solutions are given by

$$\theta = \frac{t}{\alpha_3}[p'(z) - p'(z_0)] + q(z) - q(z_0) \tag{23}$$

$$v = \frac{p(z) - p(z_0)}{(\eta_2 - \alpha_3)} \tag{24}$$

where $p(z)$ and $q(z)$ are given by

$$p(z) = k_1 e^{y_1 z} + k_2 e^{y_2 z} + p'(z_0) z, \tag{25}$$

$$q(z) = \frac{b_1 z}{y_1 + m} e^{y_1 z} - \left(z + \frac{1}{2m}\right)\frac{e^{y_2 z}}{2m} - \frac{ne^{-mz}}{2m} + Se^{mz} - \frac{C}{m^2} \tag{26}$$

and

$$y_{1,2} = \pm\sqrt{A/k_{33}},$$

$$k_1 y_1 e^{y_1 z_0} + k_2 y_2 e^{y_2 z_0} = 0$$

$$b_1 = \frac{k_l y_1 b}{\alpha_3 a}$$

$$C = -\frac{p'(z_0)}{a} - \frac{A}{k_{33}} q(z_0) - \frac{B}{2k_{33}}$$

with three constants m, n and S.

We now present the third solution to the problem in order to show the inconsistencies that arose when solving the equation using separation of variables. Setting $\theta = Z(z)T(t)$ and substituting it into Eq. 12, we have

$$a\frac{Z'''}{Z'} + c = b\frac{T'}{T} = D \tag{27}$$

where D is the separation constant. For the case in which $(c - D)/a$ is positive, Eq. 27 has a solution

$$T = T_0 e^{Dt/b} \quad Z = f_1 \cos \xi z + f_2 \sin \xi z - \frac{m}{\xi^2} \tag{28}$$

where $\xi = \sqrt{(c - D)/a}$, f_1, f_2 and m are constants. The sine and cosine terms give either the bend or splay modes. Since our solution is independent of k_{11}, all nematogenic compounds will exhibit a universal phenomenon of the splay mode for small distortions. Although some physics may be lost, we set $f_2 = 0$ in Eq. 28 with the boundary condition $\theta(z_0) = 0$ to obtain a specific solution

$$\theta = \theta_0[\cos z - \cos z_0]e^{Dt/b} \tag{29}$$

$$v = e^{Dt/b}[E(\sin \xi z - \sin \xi z_0) + F(z - z_0)] + \frac{B}{2\alpha_2}(z - z_0) \tag{30}$$

where

$$\theta_0 = T_0 f_1$$

$$E = \frac{\theta_0}{\alpha_2 \xi}\left[\xi^2 k_{33} - (\alpha_3 - \alpha_2)\frac{D}{b} - A\right]$$

$$F = \frac{\theta_0}{\alpha_2}\cos \xi z_0\left[(\alpha_3 - \alpha_2)\frac{D}{b} + A\right]$$

III. CONCLUSIONS

The purposes of this work are to reveal the inconsistencies of the solutions for the distortion waves in the Freedericks' transition problem and to give consistent and generalized solutions for replacing those existing in the literature. Three solutions for θ and v are obtained as functions of z and t. The first solution is the most general. It makes no assumptions about the form of the solution and no initial nor boundary conditions are applied until the general forms of θ and v are obtained. Because of the generality and non-uniqueness of this solution for these partial differential equations, it can be used to predict possible distortion patterns and to interpret other physical phenomenon by varying the form of the $c_i(t)$'s and the function $g(bz + t)$. Its practical applications are under investigation and will appear in the literature at a later time.

The second solution is a more specific one. It uses the basic assumption $\partial v/\partial t = 0$, which leads to the inertia torque term to vanish. The final solution remains consistent with this assumption, which implies $v = f(z)$. The final solution does indeed show v to be a function of z only. The only arbitrariness comes from choosing the form of the integration constants. Both of these solutions are good only for the state of *steady flow*. Actually, the equation $\partial v/\partial t = 0$ is not physically realizable. (It cannot be used to explain the whole process of the Freedericks' transition.) Without doing this, however, it is difficult to obtain analytical solutions. A better approach would be to set up some model for the time dependence of v which gives $\partial v/\partial t$ approximately equal to zero; i.e., v is a slowly varying function of t.

We now discuss the third solution to the problem which is obtained by separation of variables z and t, and commonly adopted in papers appearing in the literature. Clearly, these results contradict the basic assumptions of $\partial v/\partial t = 0$ and $\partial^2\theta/\partial t^2 = 0$. It is easy to show that this method forces a solution to become inconsistent with the physics involved. Eq. 10 can be integrated to give the equation

$$(\eta_2 - \alpha_3)\frac{\partial v}{\partial z} + \alpha_3\frac{\partial \theta}{\partial t} = G(t) \tag{31}$$

If this is substituted in Eq. 11, we obtain an equation of the form

$$P\frac{\partial^2\theta}{\partial z^2} + Q\frac{\partial \theta}{\partial t} + R\theta + \frac{B}{2\alpha_2} - G(t) = 0 \tag{32}$$

where P, Q, and R are functions of α_i, η_i, k_{ii}, and ϕ. Assuming $\theta = Z(z)T(t)$, we have

$$P\frac{Z''}{Z} + Q\frac{T'}{T} + R + \frac{\left[\frac{B}{2\alpha_2} - G(t)\right]}{ZT} = 0 \tag{33}$$

This means that $[(B/2\alpha_2) - G(t)]$ must equal zero or our separation of variables assumption will not be accomplished. Note that the solutions using separable variables can also be obtained by assuming $[(B/2\alpha_2) - G(t)] = KT$, where K is an arbitrary constant. However, if one substitutes $G(t)$ into Eq. 31 and takes the derivatives of Eq. 31 with respect to time, the left-hand side of the equation should vanish because of the consistency of the basic assumption that the linear and angular momenta of the fluid are conserved. Consequently, the time derivative of $G(t)$ will disappear. It then will lead again to the only solution with zero K-value. A further implication is that $G(t) = \text{constant} = B/2\alpha_2$. This is in general not true except for the states of steady flow. We have shown earlier that $G(t) = g'(z_0) = B/2\alpha_2$. Equation 29 shows that the angular velocity of the director depends on the gradient of the fluid velocity and another source which is independent of position, i.e., $g'(z_0)$.

Another problem arising from these solutions is the assumption that $\partial v/\partial t = 0$. Remember that this assumption implies $v = f(z)$. However, the solution given for v is a function of z and t except when $D = 0$. For $D = 0$, we obtain only the profile of θ, i.e., static case only.

Finally, there is also a problem with the equilibrium states, which occur only when both the angular velocity of the director and the fluid velocity gradient vanish, i.e.,

$$\frac{\partial \theta}{\partial t} = \frac{\partial v}{\partial z} = 0$$

Thus we have from Eq. 31

$$G(t) = \frac{B}{2\alpha_2} = \frac{\chi_a H^2}{2\alpha_2} \sin 2\phi = 0$$

which is not necessarily the case in an actual physical situation.

In order to find the time required to complete a Freedericks' transition, one can use Eq. 23. If we take the partial derivative of θ with respect to z and set it equal to zero, then we can solve for values of z which give the maximum distortion. As long as the maximum distortion of θ is less than 30° (small angle assumption, sin $\theta \sim \theta$), then this approach is valid. Using the value of θ_{max} from an experimental measurement and our value of z_{max}, one can solve for t from Eq. 23.

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