We conclude this comment by pointing out that the temporal and spatial formulations of linear stability of parallel flows with respect to traveling dispersive or non-disturbances are equivalent within the framework of linear theory if the conditions 1 and 2 are met. The application of the linear theory to the nonlinear wave regime only leads to confusion and does not predict wave properties (wave speed, amplitude, length and growth rate) quantitatively. Quantitative understanding of film stability must be based on nonlinear stability theories. Unfortunately, the existing nonlinear theories are rather limited in scope. For example, the theory of Lin (1969, 1970, 1974) is valid only for $\alpha_r << 1$ and comparisons of his theory with the observations of Krantz and Goren (1971) for finite values of α_r are not even possible. Finally, we mention the work of Agrawal (1972) on the nonlinear stability of liquid films with respect to spatially growing disturbances.

ACKNOWLEDGMENT

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NOTATION

C_g = group velocity
C_i = imaginary part of the complex wave velocity
C_r = real part of the complex wave velocity
h_0 = mean film thickness

Greek Letters

α_r = wave number = $2\pi h_0/\lambda$
α_i = negative of spatial amplification rate
β_i = temporal amplification rate = $\alpha_r C_i$
λ = wave length

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Additional Comments on the Spatial Formulation of the Orr-Sommerfeld Equation for Thin Liquid Films

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In the previous note Lin (1974) has presented further discussion on a recent paper of Krantz and Owens (1973). The latter developed an approximate closed-form analytical solution to the spatial formulation of the Orr-Sommerfeld equation appropriate to falling film flow. Their results, when compared with the wave property data of Krantz and Goren (1971), suggest that the spatial formulation is superior to the temporal formulation of the Orr-Sommerfeld equation for this flow. Lin is critical of these results. His principal comments concerning the work of Krantz and Owens are summarized here:

1. Lin asserts that Krantz and Owens' conclusions were based on an inappropriate use of Gaster's (1965) theorem in that the data used by Krantz and Owens involved wave properties and in particular growth rates which were dependent on the wave amplitude; that is, Lin claims that these data did not apply to the initial stages of growth. Furthermore Lin states that "... for the experimental points in Figure 5 [of Krantz and Owens (1973)] the corresponding wave amplitudes are already comparable to the film thickness, and thus the condition 2 [of Lin (1974)] is violated."

2. Lin suggests that the quantitative agreement between the data and the solution of Krantz and Owens might be an artifact of this approximate method of solution.

3. Lin maintains that the finite amplitude effect could decrease rather than increase the wave speed as suggested by Krantz and Owens. In this note we will briefly reply to each of these comments of Lin.

Lin begins by reviewing the arguments of Gaster (1965) leading to the relation between the spatial and temporal amplification rates. These arguments were omitted from the paper of Krantz and Owens but were included in the thesis of Owens (1972). This development of Lin is instructive and perhaps should have been included in the paper of Krantz and Owens. We wish to add, however, that in order to arrive at Equation (1) in Lin's note, it is necessary to assume that the disturbances

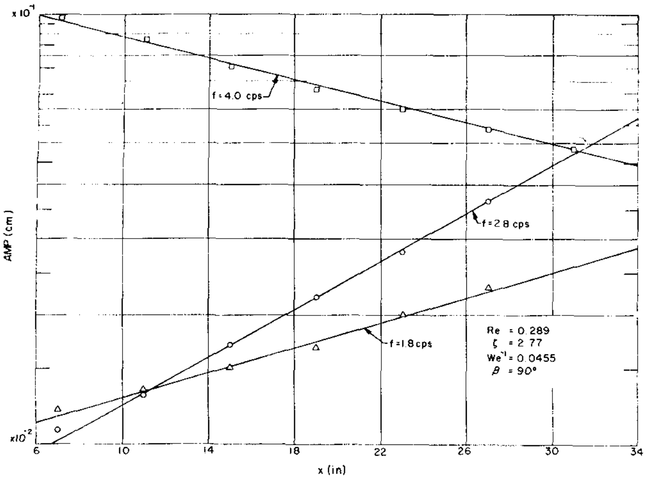

Fig. 1. Typical data showing exponential growth or decay of wave amplitude in distance.

are weakly amplified; that is, that $|c_i| << c_r$ and $|\alpha_i| << \alpha_r$. These conditions would appear to be more restrictive than condition 1 in Lin's note. Further support for these conditions comes from Gaster's (1965) original development of this equation in which he states "This method which involves an expansion is only valid for small values of amplification (or damping) and for other flows which are less stable than boundary layers it is necessary to solve the characteristic equation for each type of mode." This latter has been done for highly unstable falling film flow in Krantz and Owens (1973).

Professor Lin's comments concerning the spatial and temporal formulations have drawn our attention to some confusion which exists concerning the interpretation of the derivative $\partial(\alpha_r c_r)/\partial\alpha_r$ in Equation (1) in Lin's note. If one has a wave system, as opposed to a monochromatic wave, this derivative can be interpreted as the group velocity of the wave system. However, the concept of group velocity is somewhat devoid of meaning for a monochromatic wave. Although a monochromatic wave may have a constant phase velocity, one cannot state that this phase velocity c_r is equal to the derivative $\partial(\alpha_r c_r)/\partial\alpha_r$ as was done in Lin's note. Although a given monochromatic wave on falling film flow has a constant phase velocity, it is important to note that infinitesimal waves on this flow are dispersive; that is, a wave system is dispersive and has a group velocity given by $\partial(\alpha_r c_r)/\partial\alpha_r$. To state that this derivative is the group velocity of a monochromatic wave is to give a physical interpretation to Equation (1) in Lin's note which is only meaningful in the case of a wave system. In his original derivation of Equation (1) Gaster (1965) has stated that "... this relationship is not a physical transformation and does not imply any change in coordination system—it is just a convenient way of relating the eigenvalues in the two cases."

Lin's first criticism cited above appears to be based on a misinterpretation of the data of Krantz and Goren (1971). In the latter paper the authors state: "In all cases, waves of sufficiently small amplitude were seen to grow or decay exponentially in distance, as would be predicted for an infinitesimal wave. Exponential growth persisted until the wave amplitude approximated the film thickness or equilibrium amplitude." In addition they state that"... as long as the waves remained two-dimensional, the wave length and wave velocity remained constant, within experimental error, with distance downstream, even when amplitudes approached the equilibrium amplitude." Typical growth and decay curves used to determine the amplification rates in the experiments of Krantz and Goren are shown *in Figure* 1 as a plot of the logarithm of the wave amplitude versus streamwise distance. The basic film thickness for these data was 0.131 cm. Thus the amplification factors used in the analysis of Krantz and Owens were not determined for waves having amplitudes comparable to the film thickness as stated by Lin. Hence it would appear reasonable that the wave property data employed by Krantz and Owens can be used to test the predictions of linear stability theory.

Lin's second comment suggests that the agreement between theory and experiment shown in the work of Krantz and Owens may be accidental. Lin states that "... in the solution of Krantz and Owens, it is assumed that the parabolic velocity profile in the primary flow can be replaced by a constant." This is somewhat misleading. The solution of Krantz and Owens is based on the surface approximation of Anshus and Goren (1966) whereby the velocity profile and its derivatives are replaced by their values at the gas-liquid interface. Thus the resulting approximate form of the Orr-Sommerfeld equation contains the exact (nonzero) value of the second derivative of the velocity profile. Admittedly this solution involves an ad hoc assumption whose validity cannot be assessed a priori. One then might be apprehensive regarding any conclusion drawn from this solution concerning the relative merits of the spatial and temporal formulations. However, additional support is given to the conclusions of Krantz and Owens regarding the superiority of the spatial formulation by the recent work of Shuler (1974). Shuler developed an exact asymptotic solution to the spatial formulation of the Orr-Sommerfeld equation using the power-series method proposed by Benjamin (1957). This solution, in contrast to a similar solution to the temporal formulation, yielded quantitative agreement with the low Reynolds number data of Krantz and Goren.

The third criticism of Lin is completely justified and the authors acknowledge their error. Our statement that "the nonlinear stability theories of Nakaya and Takaki, Lin, and Gjevik predict that the wave velocity of infinitesimal waves *will* increase as the waves reach finite amplitude" should be corrected to read "*can* increase." Indeed these nonlinear theories indicate that finite amplitude effects can result in either an increase or a decrease in the wave velocity. However, since none of these investigators present any theoretical results for the White Oils used in the study of Krantz and Goren, it is impossible to infer whether finite amplitude effects can explain the small (5%) increase in the wave speed observed by Krantz and Goren.

In concluding his remarks Lin notes that the results of the spatial and temporal formulations are equivalent if his conditions 1 and 2 are met. He contends that the data of Krantz and Owens do not satisfy condition 2. Our reply to his first comment above would indicate that these data do satisfy this condition. However, Lin contends that the data of Krantz and Owens do satisfy his condition 1. We question the manner in which Lin evaluated condition 1 in that he equated the phase velocity of the monochromatic waves to the derivative $\partial(\alpha_r c_r)/\partial\alpha_r$. We claim that the spatial and temporal formulations are not equivalent for the data of Krantz and Owens because the disturbances are not weakly amplified. Indeed for Figure 3 in Krantz and Owens one finds that for the most highly amplified wave $\alpha_r \simeq 0.46$; $|\alpha_i| \simeq 0.05$; $c_r \simeq 2.6$; and $c_i \simeq 0.28$.

On the basis of the foregoing arguments we maintain that Krantz and Owens did apply the theorem of Gaster correctly and that their conclusions concerning the merits of the spatial formulation have merit. We hope that these comments will clarify any misunderstanding concerning this paper.

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New Methods for the Estimation of the Viscosity Coefficients of Pure Gases at Moderate Pressures (with Particular Reference to Organic Vapors)

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In an earlier communication (Reichenberg, 1973) it was shown that the frequently observed numerical indeterminacy of the potential parameters σ and ϵ/k is due to the fact that, under certain circumstances, these two parameters "collapse" together. When this occurs with a potential function involving only these two parameters, the predicted values of the physical property in question are effectively determined by a single parameter characteristic of the substance. The known fact that, even when this occurs, the predicted values continue to agree well with experimental values suggests that it may be possible to develop relationships which contain only a single parameter characteristic of the substance yet provide good estimates of the physical property over a wide range of temperatures.

In the particular case of the viscosity of pure gases at moderate pressures, analysis of the Lennard-Jones (12:6) potential function showed that one of the regions in which indeterminacy will occur is given by $0.4 \leq kT/\epsilon \leq 1.6$. Within this range, the Chapman-Enskog relation reduces to

$$\eta = aT \tag{1}$$

where η is the gas viscosity and a is a constant characteristic of the substance. Use may be made of the relation $k\,T_c/\epsilon \approx 1.3$ to convert these limits to $0.3 \leq T_R \leq 1.2$. Although the numerical value of kT_c/ϵ for the Lennard-Jones (12:6) function has not been completely finalized (see, for example, Barker et al., 1966), there can be no doubt that the critical temperature falls within this particular range of indeterminacy. If, therefore, within this range, Equation (1) were to hold exactly, then

$$a = \eta_c/T_c \tag{2}$$

Thus both $(\eta_c/T_c) - (\eta/T)$ and $d \log (T/\eta)/d \log T_R$ should be zero within this range. In Figure 1 it is shown that, on the contrary, a number of nonpolar gases exhibit the property that, at $T_R = 1$,

$$d \log (T/\eta)/d \log T_R = \alpha \tag{3}$$

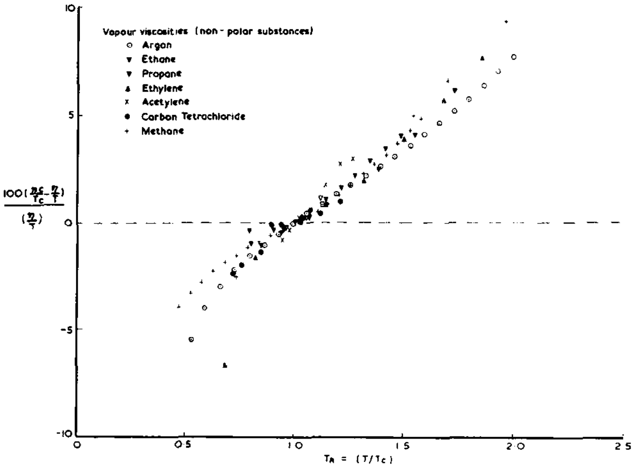

Fig. 1. Plot of 100 $(\eta_c/T_c - \eta/T) / (\eta/T)$ against T_R for nonpolar substances.