Forced Convection: IV. Asymptotic Forms for Laminar and Turbulent Transfer Rates

Mass transfer rates in laminar and turbulent nonseparated boundary layers are asymptotically expanded for small values of the diffusivity $\mathcal{D}_{AB}$, with a uniform state on the mass transfer surface. Results for heat transfer follow by analogy. The thermal or binary Nusselt number at small net mass transfer rates is given asymptotically by a generalized penetration expression

$$\langle Nu \rangle = a_{00} Pe^{1/2} + a_{01} Pe^0 + \cdots \tag{A}$$

for short times, or for boundary layers that duplicate the surface tangential motion. For flows past rigid interfaces, the long-time average of $\langle Nu \rangle$ is given asymptotically by a generalized Chilton-Colburn relation

$$\langle \overline{Nu} \rangle = b_{00} Pe^{1/3} + b_{01} Pe^0 + \cdots \tag{B}$$

in regions of nonrecirculating motion. The measurable functions a_{ij} and b_{ij} depend only on the system shape and laminar or turbulent velocity field. Formal expressions for a_{00} are given, and an expression for b_{00} in steady flows. These results agree well with data on mass transfer operations in tubes, packed beds, and fluid-fluid contactors.

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Introduction

In this paper we analyze the dependence of mass transfer rates on $\mathcal{D}_{AB}$ by constructing perturbation expansions in this parameter. The expansions are asymptotically valid for small values of $\mathcal{D}_{AB}$, whether the flow is laminar or turbulent.

The simplicity of convective diffusion in the small-diffusivity limit has prompted several analytic studies of wide classes of geometries and flows. Solutions have been found for two-dimensional flows by Lighthill (1950), Levich (1962), Acrivos (1960) and Ruckenstein (1968), and for three-dimensional flows in parts I–III of this series (Stewart, 1963; Stewart et al., 1970; Stewart and McClelland, 1983). The three-dimensional theory has been shown to agree closely with corresponding experiments in packed beds (Sørensen and Stewart, 1974) and in turbulent open-channel flows (Stewart et al., 1970).

Here we extend the asymptotic analysis to higher order and to turbulent flows with shear. The exponents and stretched variables in these expansions are identified by the method of dominant balance (Prandtl, 1904; Bender and Orszag, 1978). Detailed investigations are avoided by leaving some functions to be found from data. Explicit velocity profiles then are not needed, and the results obtained are valid for quite general flows.

Problem Description

Consider a binary fluid in laminar or turbulent flow in a one-phase region $\mathcal{V}(t)$, adjoined by mass-transfer surfaces $S_0(t)$ and inactive surfaces $S_N(t)$. The region $\mathcal{V}$ may also have inlet and exit surfaces $S_1(t)$ and $S_x(t)$ as in Figure 1. The interfaces may be fixed as in a packed-bed reactor, or some may be mobile as in a distillation column. Homogeneous chemical reactions are excluded and the physical properties are regarded as constants for a given system.

The diffusive mass flux j_A is represented here by Fick's first law (Bird et al., 1960):

$$j_A = -\rho \mathcal{D}_{AB} \nabla \omega_A \tag{1}$$

Conservation of mass gives

$$(\nabla \bullet V) = 0 \tag{2}$$

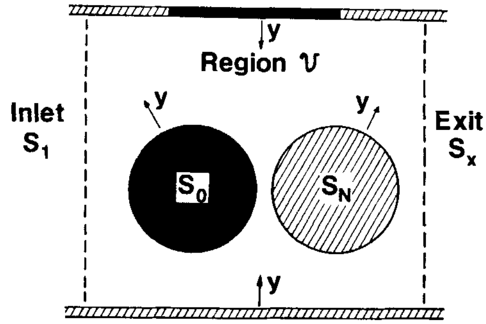

Figure 1. Generic heat or mass transfer system.
$\mathcal{V}$, one-phase region
S_0, mass transfer surfaces
S_N, inactive surfaces

for constant density, and conservation of species A gives

$$\frac{\partial \omega_A}{\partial t} + (V \bullet \nabla \omega_A) = \mathcal{D}_{AB} \nabla^2 \omega_A \tag{3}$$

for constant $\rho \mathcal{D}_{AB}$. The mass-average velocity V is measured from a stationary origin. These equations will be analyzed without smoothing; therefore, no eddy diffusivity will be needed.

The velocity field $V(r, t)$ satisfies the equation of motion, with a Newtonian or non-Newtonian expression for the stress tensor (Bird et al., 1960, 1987). For simplicity, we will not impose such restrictions on the velocity here, but will use a Taylor expansion of V in powers of the distance y from the nearest interface.

A uniform composition ω_{A1} prevails at time $t = 0$ throughout $\mathcal{V}$, and permanently in the far upstream region. Mass transfer of species A, B, or both is initiated on S_0 by a step change of interfacial composition at time $t = 0_+$. The interfacial mass fluxes are constrained, by solubility or otherwise, to a constant ratio n_{A0}/n_{B0}. Any entry and exit surfaces $S_1(t)$ and $S_x(t)$ are assumed to be far enough from S_0 that diffusion through them can be neglected. Then the initial condition at $t = 0$ is

$$\omega_A = \omega_{A1} = \text{const. in } \mathcal{V}(0) \tag{4}$$

and the boundary conditions for $t > 0$ are:

$$\omega_A = \omega_{A1} = \text{const.} \quad \text{on } S_1(t) \tag{5}$$

$$(n \bullet \nabla \omega_A) = 0 \quad \text{on } S_x(t) \tag{6}$$

$$(n \bullet \nabla \omega_A) = 0 \quad \text{on } S_N(t) \tag{7}$$

$$\omega_A = \omega_{A0} = \text{const.} \quad \text{on } S_0(t) \tag{8}$$

$$V = V_s(u, w, t) + n \frac{R_\omega \mathcal{D}_{AB}}{(\omega_{A1} - \omega_{A0})} \frac{\partial \omega_A}{\partial y} \quad \text{on } S_0(t) \text{ and } S_N(t). \tag{9}$$

Here n is the normal vector from the given surface into $\mathcal{V}$. The coordinate y of a point in $\mathcal{V}$ is the distance to the nearest interfacial element (u, w), and $V_s(u, w, t)$ is the velocity of the latter element. The mass flux ratio

$$R_\omega = \frac{(n_{A0} + n_{B0})(\omega_{A0} - \omega_{A1})}{n_{A0} - \omega_{A0}(n_{A0} + n_{B0})} \tag{10}$$

in Eq. 9 is constant in a given system; it measures the importance of the net interfacial mass flux $(n_{A0} + n_{B0})$ as in part II. The binary Nusselt number

$$\langle Nu_\omega^\bullet \rangle = \left\langle L \frac{\partial}{\partial y} \left(\frac{\omega_A - \omega_{A0}}{\omega_{A1} - \omega_{A0}} \right) \right\rangle = \langle Nu_\omega \rangle [1 + O(R_\omega)] \tag{11}$$

is to be expanded in a perturbation series for small values of $\mathcal{D}_{AB}$ and R_ω. Here L is a characteristic length of the system and $\langle\,\rangle$ denotes a surface average.

In two-phase flows each phase can be described as above, provided that Eq. 8 remains realistic. This uniform interfacial condition was derived in the small-diffusivity limit in part II, and is used here as an approximation at larger diffusivity values.

Boundary-Layer Equations

When $\mathcal{D}_{AB}$ is sufficiently small, the diffusion is localized in a boundary layer near S_0. Outside this layer, the composition is uniform. The two regions are unified by the matching condition

$$(\text{Outer limit of boundary layer composition}) = \omega_{A1} \tag{12}$$

The boundary layer solutions thus include the outer region, and satisfy Eqs. 5 and 6 asymptotically for small $\mathcal{D}_{AB}$.

The boundary layer coordinates of part II are used. Each interfacial element is permanently labeled with surface coordinates (u, w); its position vector at time t is $r_s(u, w, t)$. Each point in the boundary layer is identified by its distance, y, from the nearest interfacial point and by the surface coordinates (u, w) evaluated there. The position vector of each point (u, w, y) at time t is then expressible by a mapping function

$$r(u, w, y, t) = r_s(u, w, t) + yn(u, w, t) \tag{13}$$

relative to a stationary origin. Thus, the (u, w, y) grid moves if the interface does.

The mass-average fluid velocity relative to the origin of r is

$$V = v + (\partial r/\partial t)_{u,w,y} \tag{14}$$

Here v is the local mass-average velocity of the fluid relative to the u, w, y grid, and $(\partial r/\partial t)_{u,w,y}$ is the local velocity of this grid relative to the origin of r. Insertion of the last two relations into Eqs. 2 and 3 gives the boundary layer continuity equations

$$(\nabla \bullet v) = -(\nabla \bullet V_s) - y \frac{\partial}{\partial t} (\nabla \bullet n) \tag{15}$$

$$\left(\frac{\partial \omega_A}{\partial t} \right)_{u,w,y} + (v \bullet \nabla \omega_A) = \mathcal{D}_{AB} \nabla^2 \omega_A \tag{16}$$

in which V_s denotes $\partial r_s(u, w, t)/\partial t$, the velocity of the nearest interfacial element. The righthand terms of Eq. 15 arise from volume changes of the coordinate elements *dudwdy* near deformable interfaces. The "surface-stretch" term $(\nabla \bullet V_s)$ was treated extensively in part II of this series.

The following boundary layer operators are derived in the Appendix:

$$\nabla = n\frac{\partial}{\partial y} + \sum_{k=0}^{\infty}\left(-y\frac{\partial n}{\partial r_s}\right)^k \frac{\partial}{\partial r_s} = n\frac{\partial}{\partial y} + \sum_{k=0}^{\infty} y^k \nabla_k \quad (17)$$

$$\nabla^2 = \frac{\partial^2}{\partial y^2} + (\nabla_0 \bullet n)\frac{\partial}{\partial y} + \nabla_0^2 + \cdots \quad (18)$$

The ∇_k are tangential differential operators, and $(\nabla_0 \bullet n)$ is the sum of the principal interfacial curvatures at the point $(u, w, 0)$.

The tangential and normal components of v can be expanded in Taylor series in y to give

$$\begin{aligned} v &= v_I + n v_n \\ &= \sum_{k\geq 1} y^k[\beta_k(u, w, t) + n\gamma_k(u, w, t)] + n v_0(u, w, t) \quad (19) \end{aligned}$$

in which $n v_0(u, w, t)$ denotes the final term of Eq. 9. The functions γ_k are determined via Eq. 15, giving

$$\gamma_1 = -(\nabla_0 \bullet n)v_0 - (\nabla_0 \bullet V_s) \quad (20a)$$

$$\begin{aligned} 2\gamma_2 = &-(\nabla_0 \bullet \beta_1) - (\nabla_0 \bullet n)\gamma_1 \\ &- (\nabla_1 \bullet n)v_0 - (\nabla_1 \bullet V_s) - \frac{\partial}{\partial t}(\nabla_0 \bullet n) \quad (20b) \end{aligned}$$

$$\begin{aligned} 3\gamma_3 = &-(\nabla_0 \bullet \beta_2) - (\nabla_0 \bullet n)\gamma_2 - (\nabla_1 \bullet \beta_1) - (\nabla_1 \bullet n)\gamma_1 \\ &- (\nabla_2 \bullet n)v_0 - (\nabla_2 \bullet V_s) - \frac{\partial}{\partial t}(\nabla_1 \bullet n) \quad (20c) \end{aligned}$$

and so on. Equations 16–19 give the expanded continuity equation,

$$\begin{aligned} \left(\frac{\partial \omega_A}{\partial t}\right)_{u,w,y} &+ y(\beta_1 \bullet \nabla_0 \omega_A) + y^2(\beta_1 \bullet \nabla_1 \omega_A + \beta_2 \bullet \nabla_0 \omega_A) \\ &+ \cdots + (v_0 + y\gamma_1 + y^2\gamma_2 + \cdots)\frac{\partial \omega_A}{\partial y} \\ &= \mathcal{D}_{AB}\left[\frac{\partial^2 \omega_A}{\partial y^2} + (\nabla_0 \bullet n)\frac{\partial \omega_A}{\partial y} + \nabla_0^2 \omega_A + \cdots\right] \quad (21) \end{aligned}$$

valid for laminar and turbulent boundary layers.

Coordinate Stretching

The diffusional boundary layer may consist of several regions, as illustrated in Figures 2–5. Most of these regions shrink in one or more dimensions as the parameter $\mathcal{D}_{AB}$ is reduced. The coordinates will be stretched accordingly to express Eq. 21 in local forms that are asymptotically independent of $\mathcal{D}_{AB}$.

The main regions of diffusion lie along the interfaces, and shrink only in the y direction with $\mathcal{D}_{AB}$; see region 2 in Figures 2–5, and region 6 in Figure 2. For each such region we use a stretched coordinate

$$Y_j = y/\kappa_j(\mathcal{D}_{AB}) \quad (22)$$

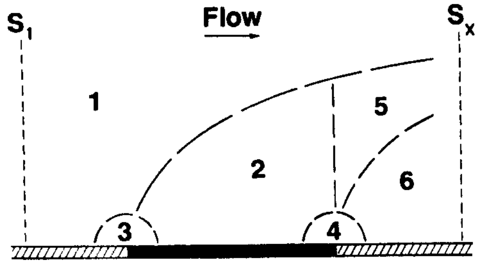

Figure 2. Fluid regions near a wall electrode (after Newman, 1973).

1. Outer region
2. Main diffusion layer
3. Leading edge region
4. Trailing edge region
5,6. Downstream regions

with a stretching function $\kappa_j(\mathcal{D}_{AB})$ to be determined. Equation 21 then takes the following form for such a region j:

$$\begin{aligned} \left(\frac{\partial \Pi_j}{\partial t}\right)_{u,w,Y_j} &+ \kappa_j Y_j(\beta_1 \bullet \nabla_0 \Pi_j) + \kappa_j^2 Y_j^2(\beta_1 \bullet \nabla_1 \Pi_j \\ &+ \beta_2 \bullet \nabla_0 \Pi_j) + \cdots + \left(\frac{\mathcal{D}_{AB} R_\omega}{\kappa_j^2}\frac{\partial \Pi_j}{\partial Y_j}\bigg|_0 \right. \\ &\left. + Y_j\gamma_1 + \kappa_j Y_j^2\gamma_2 + \kappa_j^2 Y_j^3 \gamma_3 + \cdots\right)\frac{\partial \Pi_j}{\partial Y_j} \\ &= \frac{\mathcal{D}_{AB}}{\kappa_j^2}\left[\frac{\partial^2 \Pi_j}{\partial Y_j^2} + \kappa_j(\nabla_0 \bullet n)\frac{\partial \Pi_j}{\partial Y_j} \right. \\ &\left. + \kappa_j^2 \nabla_0^2 \Pi_j + \cdots\right] \quad (23) \end{aligned}$$

in which $\Pi = (\omega_A - \omega_{A0})/(\omega_{A1} - \omega_{A0})$. To lowest order in κ_j, the operator ∇^2 of Eq. 2 takes the form $\partial^2/\partial y^2$ as in Prandtl's boundary layer equations.

Two-dimensionally shrinking boundary layer features occur along edges of S_0; also at corners and around loci of separation or of surface-surface contact; see regions 3 and 4 in Figs. 2–5. Some of these loci may be moving. For such a region we define Y_j as in Eq. 22, along with a stretched surface coordinate

$$X_j = x_j(u, w, t)/\kappa_j(\mathcal{D}_{AB}) \quad (24)$$

measured from the locus, and an unstretched surface coordinate $z_j(u, w, t)$ orthogonal to x_j. The tangential derivatives are

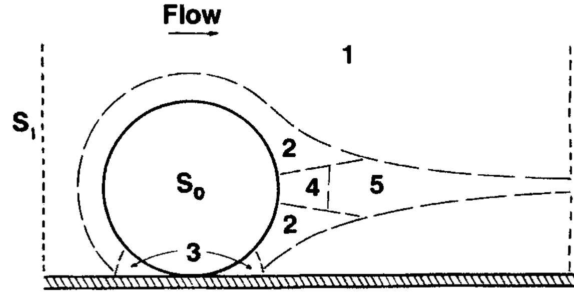

Figure 3. Fluid regions in slow flow past a sphere and inactive wall.

1. Outer region
2. Main diffusion layer
3. Fillet region
4. Separation region
5. Wake regions

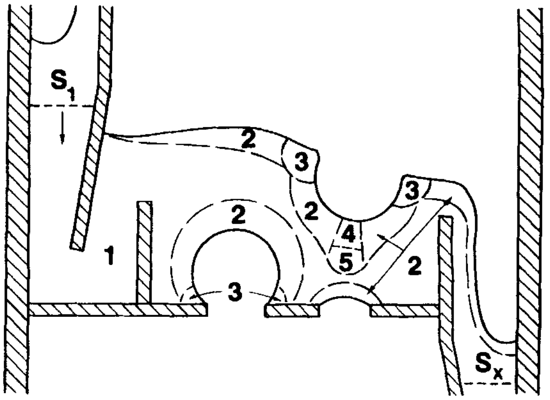

Figure 4. Liquid regions on a simple sieve tray.
1. Outer region
2. Main diffusion layer
3. Corner region
4. Separation region
5. Wake regions

rewritten accordingly with stretched operators

$$G_{kj} = \kappa_j(\mathcal{D}_{AB})\nabla_k \tag{25}$$

and Eq. 21 takes the following form for such a region j:

$$\left(\frac{\partial \Pi_j}{\partial t}\right)_{X_j,z_j,Y_j} - \frac{1}{\kappa_j}(U_j \bullet G_{0j}\Pi_j) + Y_j(\beta_1 \bullet G_{0j}\Pi_j - U_j \bullet G_{1j}\Pi_j) + \cdots + \left(\frac{\mathcal{D}_{AB}R_\omega}{\kappa_j^2}\frac{\partial \Pi_j}{\partial Y_j}\bigg|_0 + Y_j\gamma_1 + \kappa_j Y_j^2\gamma_2 + \cdots\right)\frac{\partial \Pi_j}{\partial Y_j} = \frac{\mathcal{D}_{AB}}{\kappa_j^2}\left[\frac{\partial^2 \Pi_j}{\partial Y_j^2} + \kappa_j(\nabla_0 \bullet n)\frac{\partial \Pi_j}{\partial Y_j} + G_{0j}^2\Pi_j + \cdots\right] \tag{26}$$

Here U_j is the local velocity of the X_j, z_j grid relative to the u, w grid. Such a velocity arises whenever the mapping functions x_j and z_j are time-dependent.

Additional regions with different shrinkage occur if the fluid leaves S_0; see regions 5 and 6 in Figure 2, and the wakes in region 5 of Figures 3, 4 and 5. We restrict the present analysis of $\langle Nu_\omega^\bullet \rangle$ to regions of nonrecirculating flow, whose fluid elements have never left the neighborhood of S_0; then we need not analyze the other regions.

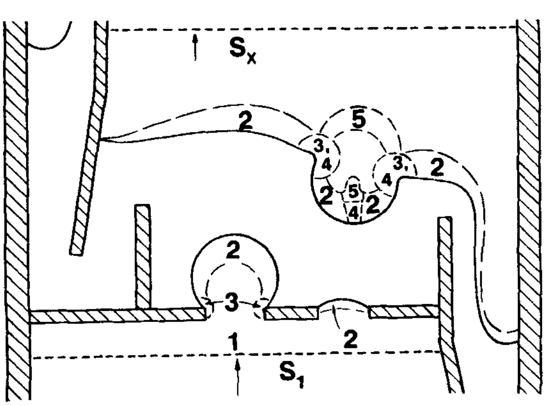

Figure 5. Gaseous regions on a simple sieve tray.
1. Outer region
2. Main diffusion layer
3. Corner region
4. Separation region
5. Wake regions

Unsteady Mass Transport for Short Times

Beginning at $t = 0$, a diffusional boundary layer propagates into $\mathcal{V}$ from S_0. Except for edge regions, the diffusion in this layer is directed essentially normal to S_0 and Eq. 23 applies, as in region 2 of Figure 2, 3, 4, or 5.

In the limit of small $\mathcal{D}_{AB}$ and small R_ω, Eq. 23 takes the asymptotic form

$$\frac{\partial \Pi_2}{\partial t} + Y_2\gamma_1\frac{\partial \Pi_2}{\partial Y_2} \sim \frac{\mathcal{D}_{AB}}{\kappa_2^2}\frac{\partial^2 \Pi_2}{\partial Y_2^2} \tag{27}$$

since positive powers of κ_2 and R_ω can then be neglected. This result holds for any finite t, but the limit is approached most rapidly when t is small. The principle of dominant balance (Bender and Orszag, 1978) requires that the dominant terms be of equal order in $\mathcal{D}_{AB}$; this gives

$$\kappa_2 = \mathcal{D}_{AB}^{1/2} \tag{28}$$

within an arbitrary multiplicative constant. Thus, the thickness of the main boundary layer region varies as $\mathcal{D}_{AB}^{1/2}$, in agreement with the detailed solutions of Eq. 27 given in part II.

Equations 23 and 28 yield a perturbation differential equation, expanded in powers of $\mathcal{D}_{AB}^{1/2}$ and R_ω. Choosing dimensionless variables

$$Z = Y_2/\sqrt{t} = y/\sqrt{\mathcal{D}_{AB}t} \tag{29}$$

$$Pe = L^2/(\mathcal{D}_{AB}t) \tag{30}$$

we find that the solution subject to Eqs. 4, 8, and 12 has the following form for any given geometry and velocity field:

$$\Pi_2 = f_{00}(u, w, Z, t) + Pe^{-1/2}f_{01}(u, w, Z, t) + R_\omega f_{10}(u, w, Z, t) + O(\epsilon^2) \tag{31}$$

Here ϵ is the array $(Pe^{-1/2}R_\omega)$. The resulting contribution to the surface-mean Nusselt number is

$$\langle Nu_\omega^\bullet \rangle_2 = \frac{L}{\sqrt{\mathcal{D}_{AB}t}}\left\langle\frac{\partial \Pi_2}{\partial Z}\right\rangle = Pe^{1/2}\left[\left\langle\frac{\partial f_{00}}{\partial Z}\right\rangle + Pe^{-1/2}\left\langle\frac{\partial f_{01}}{\partial Z}\right\rangle + R_\omega\left\langle\frac{\partial f_{10}}{\partial Z}\right\rangle + O(\epsilon^2)\right] \tag{32}$$

for systems with fixed or mobile interfaces.

The remaining fluid regions along S_0 are governed by Eq. 26. Here the appropriate form of $\kappa_j(\mathcal{D}_{AB})$ depends on the form of U_j or β_1, whichever is dominant around $x_j = 0$. In every case, the stretching will be equal in Y_j and X_j as shown in Eqs. 22 and 24. The resulting contributions to $\langle Nu_\omega^\bullet \rangle$ are at most $O(\kappa_j^0)$ =

$O(Pe^0)$, since the elements of area for surface averaging in Eq. 11 are proportional to κ_j or to κ_j^2 and the y derivative is proportional to κ_j^{-1}. Combining this result with Eq. 32 and taking the R_ω dependence from part II, we get

$$\langle Nu_\omega^\bullet \rangle = a_{00} Pe^{1/2}\left(1 - \frac{2}{\pi} R_\omega\right) + a_{01} Pe^0 + \cdots \quad (33)$$

of which Eq. A is a special case. Finally, Eq. 41 of part II yields

$$a_{00} = \frac{2}{\sqrt{\pi}} \frac{1}{A_0} \iint_{\mathcal{R}} t^{1/2} \frac{\partial}{\partial t} \sqrt{\int_{t_0(u,w)}^{t} s^2(u, w, t_1)\, dt_1}\, du dw \quad (34)$$

when the mass transfer coefficient is evaluated for a time-dependent mass transfer region $\mathcal{R}(u, w, t)$ but referred to a constant reference area A_0. Here $s(u, w, t_1)\, dudw$ is the area of the interfacial element $dudw$ at time t_1. A variable starting time $t_0(u, w)$ for the mass transfer is included here as in part II.

The new feature of Eq. 33 relative to part II is the Pe^0 term. Contributions of this order come from the velocity derivatives β_1 and γ_2, from interfacial curvature $(\nabla_0 \bullet \boldsymbol{n})$, and from edges or other regions described by Eq. 26.

Mass Transport Dominated by Interfacial Motion

Equations 27–34 remain valid for longer times if the convection in the boundary layer is dominated by the interfacial motion. This happens in regions with negligible β_1 as in laminar falling films, and in regions of positive surface stretch rate $(\nabla \bullet \boldsymbol{V}_s)$ as in laminar liquid jets.

Some of these systems become steady from an external frame of reference, although they remain time-dependent in the interfacially imbedded coordinates u, w, y. Equations 32 and 33 then can be applied more easily if we define new dimensionless variables as follows:

$$Z^{ss} = Y\sqrt{L/V_0} = y/\sqrt{\mathcal{D}_{AB} L/V_0} \quad (29a)$$

$$Pe^{ss} = VL/\mathcal{D}_{AB} \quad (30a)$$

With these changes, the coefficients in Eq. 33 become independent of time; the leading coefficient is obtainable from Eq. 84 of part II:

$$a_{00}^{ss} = \frac{1}{A_0}\sqrt{\frac{4L}{\pi V_0}} \int_{u_I}^{u_{II}} \sqrt{\int_0^{\ell(u)} \left[\frac{V_s}{(\nabla u)^2}\bigg|_{u,\ell_1}\right] d\ell_1}\, du \quad (34a)$$

Here the surface streamlines are used as u contours, and ℓ_1 is the arc length along the local u contour from the entrance of the mass transfer region.

Steady Mass Transport Near Rigid Interfaces

For steady state systems with rigid interfaces, Eq. 23 takes the asymptotic form

$$\kappa_2 Y_2 (\boldsymbol{\beta}_1 \bullet \nabla_0 \Pi_2) - \frac{1}{2}\kappa_2 Y_2^2 (\nabla_0 \bullet \boldsymbol{\beta}_1) \frac{\partial \Pi_2}{\partial Y_2} \sim \frac{\mathcal{D}_{AB}}{\kappa_2^2} \frac{\partial^2 \Pi_2}{\partial Y_2^2} \quad (35)$$

for small $\mathcal{D}_{AB}$ and small R_ω. Here Eqs. 20a and 20b have been used, and the subscript 2 denotes the main mass transfer region as in Figures 2 and 3. Dominant balance requires that κ_2 and $\mathcal{D}_{AB}/\kappa_2^2$ be proportional functions of $\mathcal{D}_{AB}$; this gives

$$\kappa_2 = (\mathcal{D}_{AB} L/B)^{1/3} \quad (36)$$

when the proportionality constant is chosen to make Y_2 simple and dimensionless. Here $B(=V_0/L)$ is a characteristic magnitude of β_1 for the system.

Equations 23 and 36 give a perturbation differential equation, expanded in powers of $\mathcal{D}_{AB}^{1/3}$ and R_ω. Defining the Peclet number as

$$Pe = \frac{BL^2}{\mathcal{D}_{AB}} \quad (37)$$

one finds that the solution subject to Eqs. 8 and 12 in a given geometry and velocity field must have the form

$$\Pi_2 = g_{00}(u, w, Y_2) + Pe^{-1/3} g_{01}(u, w, Y_2) + R_\omega g_{10}(u, w, Y_2) + O(\epsilon^2) \quad (38)$$

in which ϵ is the array $(Pe^{-1/3} R_\omega)$. The resulting contribution to the mean Nusselt number is

$$\langle Nu_\omega^\bullet \rangle_2 = \frac{L}{\kappa_2}\left\langle\frac{\partial \Pi_2}{\partial Y_2}\right\rangle = Pe^{1/3}\left[\left\langle\frac{\partial g_{00}}{\partial Y_2}\right\rangle + Pe^{-1/3}\left\langle\frac{\partial g_{01}}{\partial Y_2}\right\rangle + R_\omega\left\langle\frac{\partial g_{10}}{\partial Y_2}\right\rangle + o(\epsilon)\right] \quad (39)$$

The contributions from any regions governed by Eq. 26 are of order $Pe^{1/3} o(\epsilon)$; thus the full $\langle Nu_\omega^\bullet \rangle$ agrees with $\langle Nu_\omega^\bullet \rangle_2$ to the order shown. This point is discussed for particular geometries by Acrivos and Goddard (1965) and by Newman (1973). The g_{01} term in Eq. 39 includes interfacial curvature [through $(\nabla_0 \bullet \boldsymbol{n})$, ∇_1, and γ_3] as well as velocity-profile curvature (through β_2 and γ_3); see the terms of order κ_j^2 and $\mathcal{D}_{AB}/\kappa_j$ in Eq. 23.

Calculating the R_ω term from part I and abbreviating the integrals, we get

$$\langle Nu_\omega^\bullet \rangle = b_{00} Pe^{1/3}(1 - 0.566\, R_\omega) + b_{01} Pe^0 + \cdots \quad (40)$$

which is consistent with Eq. B. Finally, Eq. 24 of part I yields

$$b_{00} = \frac{3^{1/3}}{2\Gamma(4/3)} \frac{L^2}{A_0} \int_{z_I}^{z_{II}} \left[\int_{x_1(z)}^{x_2(z)} \sqrt{\frac{h_z \beta}{LB}} \frac{h_x}{L} \frac{h_z}{L}\, dx\right]^{2/3} dz \quad (41)$$

when the orthogonal surface coordinates used there are made dimensionless with scale factors h_x and h_z proportional to L. This result holds for a mass transfer region of unseparated flow, extending downstream from $x_1(z)$ to $x_2(z)$ and laterally between a pair of surface streamlines, z_I and z_{II}.

Turbulent Mass Transport for Long Times Near Rigid Interfaces

For the long-time analysis, we use the Fourier transform

$$\int_{-\infty}^{\infty} \exp(-i2\pi f t)\, g(\bullet, t)\, dt = \tilde{g}(\bullet, f) \quad (42)$$

as extended by Lighthill (1958) to generalized functions. Here (•) denotes arguments held constant in the integration. The derivative theorem

$$\int_{-\infty}^{\infty} \exp(-i2\pi ft) \frac{\partial}{\partial t} g(\bullet, t) = i2\pi f \tilde{g}(\bullet, f) \qquad (43)$$

and the convolution theorem

$$\int_{-\infty}^{\infty} \exp(-i2\pi ft) g(\bullet, t) h(\bullet, t)\, dt = \int_{-\infty}^{\infty} \tilde{g}(\bullet, f_1) \tilde{h}(\bullet, f - f_1)\, df_1 = \tilde{g} * \tilde{h} \qquad (44)$$

follow from Eq. 42. Bracewell (1978) gives these theorems and a pictorial dictionary of transform pairs.

Fourier transformation of Eq. 23 gives

$$i2\pi f \tilde{\Pi}_2 + \kappa_2 Y_2(\tilde{\beta}_1 * \nabla_0 \tilde{\Pi}_2) + \kappa_2^2 Y_2^2(\tilde{\beta}_1 * \nabla_1 \tilde{\Pi}_2 + \tilde{\beta}_2 * \nabla_0 \tilde{\Pi}_2) + \cdots + \left(\frac{\mathcal{D}_{AB} R_\omega}{\kappa_2^2} \frac{\partial \tilde{\Pi}_2}{\partial Y_2}\bigg|_0 + Y_2 \tilde{\gamma}_1 + \kappa_2 Y_2^2 \tilde{\gamma}_2 + \kappa_2^2 Y_2^3 \tilde{\gamma}_3 + \cdots \right) * \frac{\partial \tilde{\Pi}_2}{\partial Y_2} = \frac{\mathcal{D}_{AB}}{\kappa_2^2} \left[\frac{\partial^2 \tilde{\Pi}_2}{\partial Y_2^2} + \kappa_2 (\nabla_0 \bullet \boldsymbol{n}) \frac{\partial \tilde{\Pi}_2}{\partial Y_2} + \kappa_2^2 \nabla_0^2 \tilde{\Pi}_2 + \cdots \right] \qquad (45)$$

for the main mass transport regions along a rigid interface. In the limit of small $\mathcal{D}_{AB}$ and R_ω, this equation takes the asymptotic form

$$i2\pi f \tilde{\Pi}_2 + \kappa_2 Y_2(\tilde{\beta}_1 * \nabla_0 \tilde{\Pi}_2) - \frac{1}{2} \kappa_2 Y_2^2 (\nabla_0 \bullet \tilde{\beta}_1) * \frac{\partial \tilde{\Pi}_2}{\partial Y_2} \sim \frac{\mathcal{D}_{AB}}{\kappa_2^2} \frac{\partial^2 \tilde{\Pi}_2}{\partial Y_2^2} \qquad (46)$$

after use of Eqs. 20a and 20b. Choosing κ_2 to achieve dominant balance of convection and diffusion, we get Eq. 36 again. The corresponding range of f for dominant balance of the full Eq. 46 is of order κ_2, hence of order $\mathcal{D}_{AB}^{1/3}$. In this low-frequency range, the fluctuation period $1/f$ is consistent with a diffusional "penetration distance" $\sqrt{\mathcal{D}_{AB}/f}$ of order κ_2.

For this long-time treatment, we discard Eq. 4 and extend Eqs. 8 and 12 to all time ($-\infty < t < \infty$). The resulting boundary conditions on $\tilde{\Pi}_2$ are

$$\tilde{\Pi}_2 = 0 \quad \text{at } Y_2 = 0 \quad \text{on } S_0 \qquad (47)$$

$$\tilde{\Pi}_2 \rightarrow \delta(f) \quad \text{as } Y_2 \rightarrow \infty \quad \text{for all } (u, w) \quad \text{in } S_0 \qquad (48)$$

since the Fourier transform of the generalized function 1 is the unit impulse function $\delta(f)$. Equation 12 gives a further condition if S_0 has upstream edges with locus $u = u_1(w)$:

$$\tilde{\Pi}_2 = \delta(f) \quad \text{for } Y_2 > 0 \quad \text{on } u = u_1(w) \qquad (49)$$

The upstream direction is defined here by the time-average function $[-\overline{\beta}_1(u, w)]$ in view of the importance of the low-frequency part of $\tilde{\beta}_1$ for convective diffusion.

Insertion of Eqs. 36–37 into Eqs. 45–49 gives perturbation equations for $\tilde{\Pi}_2$, expanded in powers of $Pe^{-1/3}$ and R_ω. The time-averaged solution $\overline{\Pi}_2$ has the form

$$\overline{\Pi}_2 = \overline{\Pi}_{200}(u, w, Y_2) + Pe^{-1/3} \overline{\Pi}_{201}(u, w, Y_2) + R_\omega \overline{\Pi}_{210}(u, w, Y_2) + O(\epsilon^2) \qquad (50)$$

for a given geometry and velocity field.

Therefore, the time-averaged Nusselt number is

$$\langle \overline{Nu_\omega^\bullet} \rangle_2 = \frac{L}{\kappa_2} \left\langle \frac{\partial \overline{\Pi}_2}{\partial Y_2} \right\rangle = Pe^{1/3} \left[\left\langle \frac{\partial \overline{\Pi}_{200}}{\partial Y_2} \right\rangle + Pe^{-1/3} \left\langle \frac{\partial \overline{\Pi}_{201}}{\partial Y_2} \right\rangle + R_\omega \left\langle \frac{\partial \overline{\Pi}_{210}}{\partial Y_2} \right\rangle + o(\epsilon) \right] \qquad (51)$$

The contributions from regions governed by Eq. 26 are again of order $Pe^{1/3} o(\epsilon)$, so the mean $\langle \overline{Nu_\omega^\bullet} \rangle$ over all of S_0 is likewise given by Eq. 51. This result can be written in the form of Eq. 40:

$$\langle \overline{Nu_\omega^\bullet} \rangle = Pe^{1/3} (\overline{b}_{00} + \overline{b}_{10} R_\omega) + \overline{b}_{01} + \cdots \qquad (52)$$

The coefficients $\overline{b}_{ij}$ are measurable, and are uniquely determined by the system geometry and velocity field.

The physical effects represented in Eqs. 51 and 52 are readily identified by inspection of Eqs. 45 and 46. The leading term of Eq. 52 depends, according to Eq. 46, on the Fourier transform of the interfacial velocity derivative $\beta_1(u, w, t)$. The Pe^0 term includes effects of interfacial curvature and higher velocity derivatives. The R_ω term arises as usual from the normal interfacial velocity term in Eq. 9.

Discussion

The main results of this analysis are Eqs. 33, 40, and 52 for the binary or thermal Nusselt number. Equation 33 holds for any given interfacial motion, as long as the velocity derivatives β_1 and γ_2 are unimportant in Eq. 23. Equations 40 and 52 hold at rigid interfaces in regions of nonrecirculating flow, with small $\mathcal{D}_{AB}$ and with $t\mathcal{D}_{AB}$ at large enough that the convection induced by β_1 and/or γ_2 is a dominant effect.

Fluid-fluid mass transfer operations are commonly modeled with the leading term of Eq. 33, giving a mass transfer coefficient k_ω proportional to $\mathcal{D}_{AB}^{1/2}$ for given flow conditions. This dependence is in excellent agreement with data for nonreactive gas absorption, desorption, and distillation in packed columns and plate columns, including both gas-phase and liquid-phase transfer coefficients (Sherwood and Holloway, 1940; Gerster et al., 1958; Vivian and King, 1964; Mehta and Sharma, 1966; Vidwans and Sharma, 1967). Appropriate specifications of $t_0(u, w)$ and $s(u, w, t')$ in Eq. 34 reproduce the mass transfer predictions of Ilkovic (1934), Higbie (1935), Danckwerts (1951), Cullen and Davidson (1957), Levich (1962), Chan and Scriven (1970), and others for various geometries and interfacial motions.

A stringent test of Eqs. 33 and 34 is provided by the gas absorption data of Fortescue and Pearson (1967) for open-channel flows with large eddies generated by passage through a regular grid. Insertion of their velocity distribution model into Eq. 34a allows a prediction of $\langle Nu_\omega \rangle$ based on the leading term of

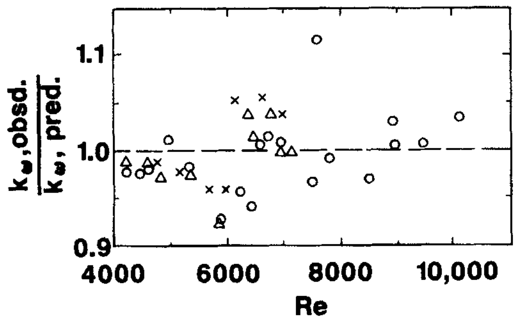

Figure 6. Turbulent gas absorption tests in an open channel compared with theory of Stewart et al. (1970).
Data from Fortescue and Pearson (1967)
Predictions correspond to the leading term of Eq. 33, with a_{00} from Eq. 34

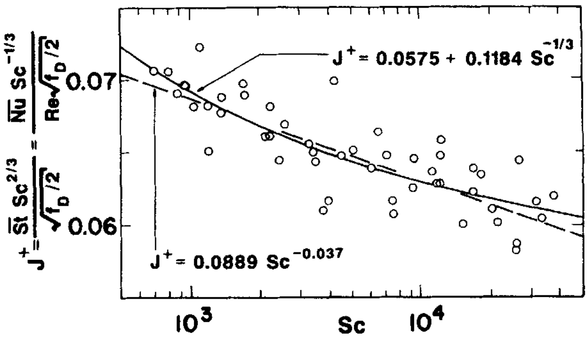

Figure 7. Correlations of data of Shaw and Hanratty (1977a) for turbulent mass transfer in pipes.
---- Shaw and Hanratty fitted power function
— fitted two-term version of Eq. 52

Eq. 33. This prediction was given in Eq. 103 of Stewart et al. (1970), and is plotted with the data in Figure 6. The agreement of the data with the theory is good, and supports (Eqs. 33 and 34 as descriptions of turbulent forced convection at mobile interfaces. The theory represents these data, and the others cited above, without postulating a surface renewal process.

The full Eq. 33 is recommended for systems with moderate Peclet numbers and for those with significant flux ratios R_ω. The Pe^0 term appears in an exact solution for the solid sphere (Carslaw and Jaeger, 1959; Sherwood et al., 1975) and in detailed theories of polarography (Koutecky, 1953; Newman, 1967) and laminar liquid jets (Duda and Vrentas, 1968). The trend of $\partial \ln k_\omega / \partial \ln \mathcal{D}_{AB}$ from 0.5 toward 1.0 with increasing stirrer speed, reported by Kozinski and King (1966) for agitated vessels, suggests that the Pe^0 term may be important for these systems also.

Equation 40 is well established for steady forced convection in laminar nonseparated boundary layers on rigid interfaces. Literature surveys are given by Newman (1973) and Stewart (1978). Expansions of higher order are available for various geometries, but merit careful examination to see if the relevant edge and separation effects have been included (Smyrl and Newman, 1971; Newman, 1973).

A three-dimensional test of Eq. 40 is provided by the electrochemical data of Karabelas et al. (1971) on mass transfer to a sphere in a dense cubic array under creeping flow conditions. They found (with $L = D$ and $B = \langle v \rangle / D$):

$$\langle Nu_m \rangle = 4.58 Pe^{1/3} \qquad \text{(Observed for } Re < 0 \text{ with } Pe > 800) \qquad (53)$$

Sørensen (1970) had independently calculated

$$\langle Nu_m \rangle = 4.50 Pe^{1/3} \text{(Predicted for } Re \rightarrow 10 \text{ with } Pe \gg 1) \qquad (54)$$

using Eq. 41 and the leading term of Eq. 40. This calculation, and a similar one for a suitably oriented simple cubic array, agreed well with the mass transfer correlation of Wilson and Geankoplis (1966) for spheres packed by dumping; see Sørensen and Stewart (1974).

Equation 52, for turbulent transport at rigid interfaces, is a natural generalization of Eq. 40 and is well supported by experiments. The leading term is consistent with the j-factor correlations of Chilton and Colburn (1934) and later workers, which have proved useful for many geometries as documented by Sherwood et al. (1975). The first two terms give an excellent fit of the data of Shaw and Hanratty (1977a) for mass transfer in tubes at $R_\omega \ll 1$ and Schmidt numbers from 693 to 37,200, as shown in Figure 7.

Various other formulas for $\langle Nu \rangle$ have been derived by eddy diffusivity approaches; the results depend on the postulated form of the eddy diffusivity function. Sandall and Hanna (1979) derived a result rather similar to Eq. 52 from an eddy diffusivity of the form $K_3 y^{+3} + K_4 y^{+4}$. The present approach is more direct, and identifies physical causes for each term in the resulting expansion.

Shaw and Hanratty (1977b) measured power spectra of the mass transfer fluctuations in turbulent flow through tubes, and found the mean frequency f_{av} to be proportional to $\mathcal{D}_{AB}^{0.404}$. Our dominant-balance analysis of Eq. 46 gives $f = O(\mathcal{D}_{AB}^{1/3})$ as the frequency range of convective diffusion. This is reasonable agreement. Further comparisons can be made once the frequency dependence of $\tilde{\Pi}_2$ is calculated.

Conclusion

Formal expansions of the Nusselt number as a function of Peclet number Pe and flux ratio R have been found here for laminar and turbulent boundary layers, by dominant balance and perturbation methods. Only minimal assumptions were made about the flow field; thus the results are applicable to many practical systems.

Regions of recirculating flow have been omitted here. These can be important, and may be treated at another time.

Acknowledgment

This research was supported by Grant NO. CPE-8308748 from the National Science Foundation, and by Grant No. DE-FG02-84ER13291 from the United States Department of Energy, Office of Basic Energy Sciences.

Notation

a_{ij}, b_{ij} = functions, Eqs. 33, 40 and 52
B = V_0/L, characteristic magnitude of β_1, s^{-1}
$\mathcal{D}_{AB}$ = binary diffusivity, $m^2 \cdot s^{-1}$
f = frequency parameter, Eq. 42, s^{-1}
f_D = friction factor
G_{kj} = stretched differential operators, Eq. 25

j_A = mass flux of species A relative to V, kg · m^{-2} · s^{-1}
j_{A0} = $\boldsymbol{n}$ component of j_A on $\mathcal{V}$ side of S_0
$k^\bullet_\omega$ = mass transfer coefficient, $j_{A0}/(\omega_{A0} - \omega_{A1})$, corrected for v_0
k_ω = limit of $k^\bullet_\omega$ as $v_0 \rightarrow 0$
L = characteristic length, m
$\langle Nu^\bullet \rangle$ = Nusselt number for diffusion, Eq. 11, or for heat transfer
$\langle Nu \rangle$ = limit of $\langle Nu^\bullet \rangle$ as $v_0 \rightarrow 0$
$\boldsymbol{n}$ = interfacial unit normal vector, directed into $\mathcal{V}$
n_{A0}, n_{B0} = mass fluxes of species A and B into $\mathcal{V}$ at $y = 0$ relative to the interface, kg · m^{-2} · s^{-1}
$O(\epsilon^k)$ = function absolutely less than $|A\epsilon^k|$ for some finite A as $\epsilon \rightarrow 0$
$o(\epsilon^k)$ = function absolutely less than $|A\epsilon^k|$ for every nonzero A as $\epsilon \rightarrow 0$
Pe = Peclet number; see Eqs. 30 and 37 for diffusional forms
R = flux ratio for diffusion, Eq. 10, or for heat transfer
$\boldsymbol{r}$ = position vector, Eq. 13, m
S_0 = mass-transfer or heat-transfer surfaces of $\mathcal{V}$
S_N = impermeable surfaces of $\mathcal{V}$
S_1, S_x = inlet and outlet surfaces of $\mathcal{V}$
Sc = Schmidt number
St = Stanton number, $NuPe^{-1}$
t = time, s
U_j = difference of grid velocities, Eq. 26, m · s^{-1}
u, w = imbedded interfacial coordinates
V_0 = characteristic velocity, m · s^{-1}
V = mass-average fluid velocity relative to origin of $\boldsymbol{r}$, m · s^{-1}
$\boldsymbol{v}$ = mass-average fluid velocity relative to u, w, y grid
$\boldsymbol{v}_\parallel$ = tangential velocity, $\boldsymbol{v} - \boldsymbol{n}v_n$
v_n = $\boldsymbol{n}$ component of $\boldsymbol{v}$
v_0 = value of v_n at $y = 0_+$; multiplier of $\boldsymbol{n}$, Eq. 9
$\mathcal{V}$ = one-phase region selected for analysis
X_j = stretched surface coordinate, Eq. 24
x_j = unstretched surface coordinate, Eq. 24
Y_j = stretched y coordinate, Eq. 22
y = distance from nearest interfacial element, m
z_j = unstretched surface coordinate orthogonal to x_j

Greek letters

β_k = coefficient of y^k in Taylor expansion of $\boldsymbol{v}_\parallel$, Eq. 19
γ_k = coefficient of y^k in Taylor expansion of v_n, Eq. 19
κ_j = stretching function, Eq. 23 or 26
Π_j = dimensionless function, $(\omega_A - \omega_{A0})/(\omega_{A1} - \omega_{A0})$ or thermal analog, for region j
ρ = density, kg · m^{-3}
ω_A = mass fraction of species A

Mathematical operations

$\langle \ \rangle$ = surface mean over S_0
$\tilde{g}$ = Fourier transform of g, Eq. 42
$\overline{g}$ = time average of g
dg_1 = dummy differential
$\tilde{g} * \tilde{h}$ = convolution of $\tilde{g}$ and $\tilde{h}$, Eq. 44
∇ = $\partial/\partial \boldsymbol{r}$
∇_k = coefficient of y^k, Eq. 17
$\sim$ = asymptotic equality

Appendix: Spatial Operators

Expansion of the operator $\nabla = \partial/\partial \boldsymbol{r}$ into normal and tangential components gives

$$\nabla = \boldsymbol{n}\left(\frac{\partial}{\partial y}\right)_{u,w,t} + \left(\frac{\partial}{\partial \boldsymbol{r}}\right)_{y,t} \tag{A1}$$

The tangential component is desired in terms of derivatives with respect to $\boldsymbol{r}_s(u, w)$. The chain rule gives

$$\left(\frac{\partial}{\partial \boldsymbol{r}}\right)_{y,t} = \left(\frac{\partial \boldsymbol{r}_s}{\partial \boldsymbol{r}}\frac{\partial}{\partial \boldsymbol{r}_s}\right)_{y,t} \tag{A2}$$

and Eq. 13 yields

$$\left(\frac{\partial \boldsymbol{r}}{\partial \boldsymbol{r}_s}\right)_{y,t} = \boldsymbol{I} + y\left(\frac{\partial \boldsymbol{n}}{\partial \boldsymbol{r}_s}\right)_t \tag{A3}$$

Inversion of this second-order matrix equation gives a geometric series, or Neumann expansion (Rall, 1969)

$$\left(\frac{\partial \boldsymbol{r}_s}{\partial \boldsymbol{r}}\right)_{y,t} = \sum_{k=0}^{\infty}\left(-y\frac{\partial \boldsymbol{n}}{\partial \boldsymbol{r}_s}\right)^k \tag{A4}$$

valid within the convergence region

$$|y\lambda_i| < 1 \quad \text{for } i = 1, 2 \tag{A5}$$

The eigenvalues λ_i of the matrix $[-\partial \boldsymbol{n}/\partial \boldsymbol{r}_s]$ are the principal curvatures of the interface at $\boldsymbol{r}_s$ (McConnell, 1957).

Insertion of Eqs. A2 and A4 into Eq. A1 gives Eq. 17, and evaluation of $(\nabla \bullet \nabla)$ gives Eq. 18.

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Manuscript received Nov. 5, 1986, and revision received June 1, 1987.