Structure and the Reflectionless/Refractionless Nature of Parabolic Diffusion-Wave Fields

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We show the impossibility of reflection and refraction phenomena at linear diffusion-wave-field (DWF) interfaces. Instead, interfacial flux expressions are derived which involve coherent accumulation or depletion phenomena subject to an interface flux conservation principle. The conditions for reflectionless and refractionless interfaces are the parabolic nature and the concomitant Fickian constitutive relations satisfied by DWFs. Simulations show that the reflection and Snell's laws can be adequate approximations only under near-normal incidence conditions, in agreement with published experimental evidence in wide areas of biomedical, electronic, and materials physics.

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In recent years the class of diffusion-based oscillations named diffusion-wave fields (DWFs) [1] has been receiving much attention [1,2]. Applications are rapidly being developed spanning thermal waves, physical electronics (carrier plasma diffusion waves), (biomedical) physics of turbid media (diffuse photon density waves), as well as mass-transport waves in stratified media, electrolytes, membranes, and polymers, and even controversial atmospheric diffusive viscosity waves [3]. Formally, diffusion waves arise from the classical parabolic diffusion equation with an oscillatory force function in homogeneous media $\nabla^2\Psi(\boldsymbol{r},t) - \frac{1}{D_k}\frac{\partial}{\partial t}\Psi(\boldsymbol{r},t) + H(\boldsymbol{r})\Psi(\boldsymbol{r},t) = \frac{1}{2}q(\boldsymbol{r})(1 + e^{i\omega t})$. Here D_k is a transport property of the medium, usually a field diffusivity in m^2/s. The driving force generates oscillatory solutions for the field (wave) function $\Psi(\boldsymbol{r},t) = \Phi(\boldsymbol{r},\omega)e^{i\omega t}$. A sort of pseudowave Helmholtz equation is thus obtained via Fourier transformation of $\Psi(\boldsymbol{r},t)$:

$$\nabla^2\Phi(\boldsymbol{r},\omega) - \kappa^2(\boldsymbol{r},\omega)\Phi(\boldsymbol{r},\omega) = Q(\boldsymbol{r},\omega), \qquad (1)$$

where $\kappa(\boldsymbol{r},\omega)$ is the complex diffusion wave number. For thermal waves $H(\boldsymbol{r}) = 0$, and $\kappa(\omega) = (1 + i)/L(\omega)$, where $L(\omega) = \sqrt{2D_t/\omega}$ is the penetration depth, known as "thermal diffusion length" and D_t is the thermal diffusivity. For other common DWFs $H(\boldsymbol{r}) = H_0$ (a constant), and the real and imaginary parts of the wave number are unequal, a fact that has important consequences in the spatial distribution of the wave field [2]. In numerous studies of boundary-value problems involving the diffusion-wave equation (1), almost invariably a propagating, traveling-wave approach is assumed in analyzing data and constructing theoretical models [4]. Central to this approach is the existence of reflection and refraction phenomena at interfaces. In fact, thermal-wave mathematical formalisms of thin multilayers to date are based on signal interpretations founded on "thermal ray" reflections at interfaces [5]. These theories have had success in support of experimental results obtained almost always under normal incidence. Recently, Almond and Patel [4]and Bertolotti *et al.* [6] have introduced three-dimensional theoretical treatments of thermal waves, which imply wave-front propagation and spatial directionality. Owing to the similarity of the parabolic diffusion-wave equation (1) to the conventional hyperbolic Helmholtz equation with a complex wave number, these treatments assume that diffusion waves act like, for example, acoustic or optical waves, thus obeying the familiar interfacial equal-angle reflection and Snell's refraction laws. O'Leary *et al.* [4] have presented extensive experimental studies of diffuse photon-density-wave fields in turbid (intralipid) media across optical interfaces which are only partly supported by theoretical interpretations based on three-dimensional coherent wave fronts reflecting and refracting at interfaces. These authors invariably noted that the wave fronts become quite distorted when the source "ray angle" exceeds ~30°. They assigned the deviations to internal reflections, diffraction, aberrations, and "spurious boundary effects." The same group (O'Leary *et al.* [4]) has further labeled diffuse photon density waves as "traveling waves." Such assignations are abundant in the literature. They are, however, inconsistent with the unidirectionality of the diffusion equation, where power propagates only according to Fourier (or Fickian) flux constitutive equations. Understanding the precise physical nature of parabolic diffusion waves thus becomes paramount, as it ultimately controls the limits of spatial resolution of multilayer structures, imaging, and tomographic reconstruction processes using these waves (subsurface mechanical defects, electronic carrier lifetime scans, and/or tumor localization).

We assume a general second order differential operator $L[u] \equiv A\frac{\partial^2 u}{\partial t^2} + B\frac{\partial^2 u}{\partial x\partial t} + C\frac{\partial^2 u}{\partial x^2} + D\frac{\partial u}{\partial t} + E\frac{\partial u}{\partial x} + Fu$, which represents the partial differential equation $L[u] = 0$. For simplicity, a one-dimensional spatial coordinate is considered. If $L[u]$ is parabolic with $A = 0$, using the parametric representation $\xi = \xi(x,t)$; $\eta = \eta(x,t)$ results in the normal form

$$L_p[u] = C\left(\frac{\partial\eta}{\partial x}\right)^2\frac{\partial^2 u}{\partial\eta^2} + D\left(\frac{\partial\xi}{\partial t}\right)\frac{\partial u}{\partial\xi} + \left[D\left(\frac{\partial\eta}{\partial t}\right) + E\left(\frac{\partial\eta}{\partial x}\right)\right]\frac{\partial u}{\partial\eta} + Fu. \qquad (2)$$

In what amounts to renormalizing the normal form above, one may set $\partial\eta/\partial x = 1$ and $\partial\xi/\partial t = 1$, in order to conform to the structures of the diffusion equations from which the commonly encountered DWFs are derived through a linear Fourier transformation. The single characteristic curve $\xi(t) = t = \text{const}$ with propagation velocity $v = dx/dt = \infty$ indicates that the parabolic field $L_p[u] = 0$ evolves unidirectionally (forward) in time away from the source everywhere simultaneously, as the operation $t \rightarrow -t$ does not leave the solution invariant. This type of simultaneity, and the existence of a single characteristic curve, precludes reflections at interfaces. The condition $A = 0$ is not central to the existence of reflectionless and refractionless interfaces. The crucial conditions for this behavior are the parabolic nature and the gradient-driven constitutive relation of the wave field. The lack of wave-front structure from parabolic diffusion fields with $A = 0$ was first recognized in a statement by Morse and Ingard in 1968 ([7], p. 479). From an intuitive viewpoint, since energy propagation is Fickian, parabolic fields must be reflectionless because energy cannot propagate back against the field gradient (particle or mass density, temperature, etc.).

Since hyperbolic wave existence hinges on the assumption of propagating wave fronts and nonstationary direction wave vector, a different physical picture must be sought at a diffusive interface. To reveal the physical aspects of interfacial relationships in diffusion-wave fields and not be hindered by the mathematics, we consider the case of thermal diffusion waves with the simplest of all possible three-dimensional sources in a thermally isotropic half-space $0 \leq z < \infty$ of thermal conductivity k_1 undergoing boundary interactions along the plane $z = 0$. The geometry involving two half-spaces separated by a plane interface is shown in Fig. 1. A point source of strength Q_0, located at $\boldsymbol{r} = \boldsymbol{r}_1$, generates thermal waves at angular modulation frequency ω. The diffusion of these waves is not wave-vector directional, but the flux $\boldsymbol{F}_j$ is solely driven by temperature gradients (Fourier's or Fick's law), $\boldsymbol{F}_j(\boldsymbol{r}, \omega) = -k_j \nabla T_j(\boldsymbol{r}, \omega)$. The spatial distribution of the thermal-wave field is proportional to the Green function with source coordinate $\boldsymbol{r}_1{:}(x_1, y_1, z_1)$ in the domain $z > 0$ (subvolume V_1 with thermal properties k_1, D_{t1}). An appropriate spatial impulse-response function must be determined in the domain $z < 0$ (subvolume V_2 with thermal properties k_2, D_{t2}). The solution is given by [1] $T_1(\boldsymbol{r}; \omega) = (D_{t1}/k_1) \iiint_{V_1} Q_0 \delta(\boldsymbol{r}_0 - \boldsymbol{r}_1) G(\boldsymbol{r} \,|\, \boldsymbol{r}_0; \omega)\, dV_0$. When applied to Eq. (1) subject to thermal-wave field and flux continuity at the interface $z = 0$, it can be expressed in terms of Hankel transforms due to the azimuthal symmetry (isotropicity) of the problem:

$$T_1(\boldsymbol{r}; \omega) = \frac{Q_0}{4\pi k_1} \int_0^\infty \frac{1}{s_1(\lambda)} \left[e^{-s_1|z - z_1|} + \Gamma_{12}(\lambda) e^{-s_1(z + z_1)} \right] J_0(\lambda\rho) \lambda d\lambda; \qquad z \geq 0$$

and

$$T_2(\boldsymbol{r}; \omega) = \frac{Q_0}{2\pi k_1} \int_0^\infty \frac{e^{-(s_1 z_1 + s_2|z|)}}{s_1(\lambda)[1 + \xi_{21}(\lambda)]} J_0(\lambda\rho) \lambda d\lambda; \qquad z \leq 0$$

on the other half-space, where $s_1(\lambda) \equiv \sqrt{\lambda^2 + \frac{i\omega}{D_{t1}}}$; $s_2(\lambda) \equiv \sqrt{\lambda^2 + \frac{i\omega}{D_{t2}}}$; $\Gamma_{12}(\lambda) \equiv \frac{\xi_{12}(\lambda) - 1}{\xi_{12}(\lambda) + 1}$; $\xi_{ij}(\lambda) \equiv \frac{k_i s_i(\lambda)}{k_j s_j(\lambda)}$, and $\rho = \sqrt{(x - x_1)^2 + (y - y_1)^2}$ is a position-vector magnitude. $J_0(\lambda\rho)$ is the Bessel function of the first kind of order zero. The diffusion-wave flux vectors in both half-spaces $j = 1, 2$ are $\boldsymbol{F}_j(\boldsymbol{r}, \omega) = -k_j \left(\frac{\partial T_j(\rho, z)}{\partial \rho} \boldsymbol{e}_\rho + \frac{\partial T_j(\rho, z)}{\partial z} \boldsymbol{e}_z \right)$ and the azimuthal symmetry of the media results in the flux vector being independent of the direction $\boldsymbol{e}_\phi$. $\Gamma_{12}(\lambda)$ can be interpreted as the "thermal-wave interface-interaction coefficient." Each term multiplying this coefficient is, therefore, associated with the value of the flux *after* the interaction of the thermal wave with the interface. In the literature this has been labeled the "reflected" component [2,4]. At any point on the interface $z = 0$ three flux vectors can be determined: incident, $\boldsymbol{F}_i$, interface-interacted, $\boldsymbol{F}_r$, and transmitted, $\boldsymbol{F}_t$; see Fig. 2. It can be shown that there exists a radial (tangential) flux discontinuity across the interface $z = 0$. If $(k_1, D_{t1}) > (k_2, D_{t2})$, or, equivalently, $e_1 > e_2$ ($e_j = k_j/\sqrt{D_{tj}}$: thermal effusivity in $J/m^2 s^{1/2} K$) the amount of discontinuity is given by

$$\boldsymbol{e}_\rho \cdot [(\boldsymbol{F}_i + \boldsymbol{F}_r) - \boldsymbol{F}_t] = \frac{Q_0}{2\pi} \left(\frac{k_1}{k_2} - 1 \right) \int_0^\infty \frac{e^{-s_1 z_1}}{s_2(\lambda)[1 + \xi_{12}(\lambda)]} J_1(\lambda\rho) \lambda^2 d\lambda. \tag{3}$$

The radial discontinuity disappears *only* if the thermal conductivities of both domains V_1 and V_2 are equal. Furthermore, conservation of diffusion-wave flux at $z = 0$ requires that

$$\boldsymbol{F}_i(x, y, 0; \omega) + \boldsymbol{F}_r(x, y, 0; \omega) = \boldsymbol{F}_t(x, y, 0; \omega). \tag{4}$$

Considering the z components of the various fluxes at the interface and noting from Fig. 2 that $\tilde{\mathbf{n}} = -\boldsymbol{e}_z$, the outward flux normal to the plane $z = 0$ is subject to the continuity condition

$$\tilde{\mathbf{n}} \cdot \boldsymbol{F}_i(x, y, 0; \omega) = \tilde{\mathbf{n}} \cdot [-\boldsymbol{F}_r(x, y, 0; \omega) + \boldsymbol{F}_t(x, y, 0; \omega)] = \frac{Q_0}{4\pi} \int_0^\infty e^{-s_1 z_1} J_0(\lambda\rho) \lambda d\lambda. \tag{5}$$

The conservation law Eq. (5) with $\mathrm{Re}\Gamma_{12}(0) > 0$ is one of *thermal-wave energy accumulation* within the domain V_1, corresponding to the thermal effusivity relation $e_1 > e_2$. If $\mathrm{Re}\Gamma_{12}(0) < 0$, i.e., in the case $e_1 < e_2$, the opposite occurs and Eq. (5) becomes a *thermal-wave energy depletion*

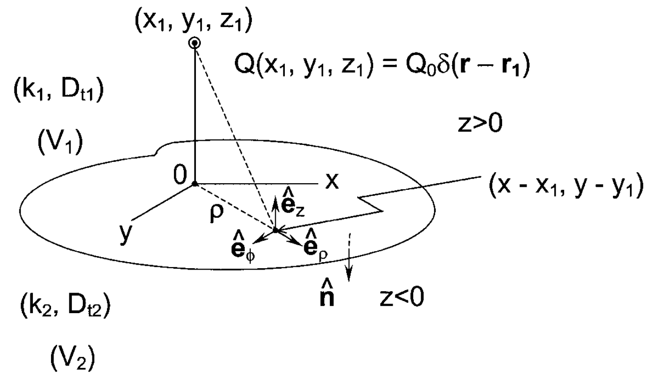

FIG. 1. Geometry of an infinite plane interface ($z = 0$) separating two half-spaces, one with a point source of diffusion waves at $\boldsymbol{r} = \boldsymbol{r}_1$.

condition. Using the vector components $\tilde{\mathbf{n}} \cdot \boldsymbol{F}_i \equiv F_{iz}, \tilde{\mathbf{n}} \cdot \boldsymbol{F}_r \equiv F_{rz}, \tilde{\mathbf{n}} \cdot \boldsymbol{F}_t \equiv F_{tz}$, Fig. 2, the continuity condition of normal flux across the interface, Eq. (5), can be written in terms of the norms $\|F_{iz}\| = \|F_{rz}\| + \|F_{tz}\|$ in the accumulation case, i.e., when $\mathrm{Re}\Gamma_{12}(0) > 0$. On the other hand, if $k_1 < k_2$ and $e_1 < e_2$, the coefficient $\mathrm{Re}\Gamma_{12}(0) < 0$, and the vector $\boldsymbol{F}_r$ points into the domain $z < 0$.

The continuity condition of normal flux across the interface, Eq. (5), can be written $\|F_{iz}\| + \|F_{rz}\| = \|F_{tz}\|$. From the interface vector diagram of Fig. 2, the incidence and interface-interacted angles can be calculated from $\tan\theta_1 = |\int_0^\infty \frac{e^{-s_1 z_1}}{s_1(\lambda)} J_1(\lambda\rho)\lambda^2\, d\lambda| \,/\, |\int_0^\infty e^{-s_1 z_1} J_0(\lambda\rho)\lambda\, d\lambda|$ and $\tan\theta_2 = |\int_0^\infty \frac{e^{-s_1 z_1}}{s_1(\lambda)} \Gamma_{12}(\lambda) J_1(\lambda\rho)\lambda^2\, d\lambda| / |\int_0^\infty e^{-s_1 z_1}\Gamma_{12}(\lambda) J_0(\lambda\rho)\lambda\, d\lambda|$. The amplitudes must be used instead of the real parts, as a fraction of a given DWF flux vector is stored in the out-of-phase component and must be accounted for. It is clear that for a given radial location ρ in general $\theta_1 \neq \theta_2$, with the exception of $\Gamma_{12}(\lambda) = 1$. This can occur only when $\xi_{12}(\lambda) \to \infty$, i.e., when $k_2 = 0$ (the medium in the subvolume V_2 is a perfect insulator). This leads to the *perfect accumulation condition:* $\boldsymbol{F}_1(\rho, 0) = \mathbf{0}$. It occurs because the flux conservation law Eq. (4) at the interface $z = 0$ reduces to $\boldsymbol{F}_r(\rho, 0) = -\boldsymbol{F}_i(\rho, 0)$. This implies $|\theta_1| = \theta_2$ in Fig. 2, reminiscent of the conventional reflection law. For all other interface-interaction coefficients, that law is not valid. For $\Gamma_{12}(\lambda) = -1$, implying $\xi_{12}(\lambda) = 0$, i.e., $k_2 \to \infty$, a physically different picture emerges, although $\tan\theta_1 = \tan\theta_2$. Here, the flux vector $\boldsymbol{F}_r$ points in the direction opposite to the one shown in Fig. 2, so that $\boldsymbol{F}_i(\rho, 0) + \boldsymbol{F}_r(\rho, 0) = \mathbf{0}$, yielding again $\boldsymbol{F}_1(\rho, 0) = \mathbf{0}$. This occurs so that the flux will remain bounded across the interface, on the other side of which it diverges, $\boldsymbol{F}_t \to \infty$. The angular relationship resulting from the equality of tangents is $|\theta_1| = \pi - \theta_2$. We may now examine the case of Snell's law of refraction. Using elementary trigonometric identities

$$\tan\theta_3 = \left| \int_0^\infty \frac{e^{-s_1 z_1}}{s_2(\lambda)[1 + \xi_{12}(\lambda)]} J_1(\lambda\rho)\lambda^2\, d\lambda \right| \Bigg/ \left| \int_0^\infty \frac{e^{-s_1 z_1}}{[1 + \xi_{12}(\lambda)]} J_0(\lambda\rho)\lambda\, d\lambda \right| . \quad (6)$$

It follows that $(\sin\theta_3/\sin\theta_1) = f(\rho)$, where $f(\rho)$ is a complicated function of the radial distance along the plane $z = 0$. This equation negates Snell's law of refraction for DWFs, according to which the ratio of the sines of the incident and transmitted/refracted angles must be constant for any incident angle θ_1; i.e., it must be independent of radial position ρ. This result is consistent with the absence of propagating coherent wave-front structure in the diffusion-wave field. Figure 3 shows plots of the relation of $|\theta_1|$ to θ_2 corresponding to $\Gamma_{12}(0) > 0$ (accumulation), and the special case $\Gamma_{12}(\lambda) = 0$, $(e_1 = e_2)$ which, however, does *not* imply identical k and D_t across the interface $z = 0$. These simulations show that under interfacial accumulation conditions the departure from the reflectionlike behavior (45° line) occurs for $\theta_2 > 30°–40°$ for most Γ_{12} cases other than zero. This is consistent with experimental reports using diffuse photon density waves (O'Leary *et al.* [4]). The greater the transport property discontinuity across the interface, the more reflectionlike the behavior is (strong accumulation limit). Extensive simulations reveal that the zigzags across the 45° line at higher angles are associated with interferencelike (linear superposition) phenomena between the real and imaginary components of the coherently diffusing power. Simulations of Eq. (6) have shown that, under accumulation, Snell's law is approximately valid for near-normal incidence [small distances ρ away from the point $(0, 0)$ on the $z = 0$ plane] and adiabatic interfaces, with the transport behavior deviating significantly at longer distances. This occurs for the same reasons as discussed with regard to Fig. 3. Under depletion, deviations appear at all distances away from the point $(0, 0)$; see Fig. 4.

To fully elucidate the causes of the accumulation/depletion behavior of diffusion waves at interfaces, it is useful to compare it with that of conventional scalar hyperbolic propagating wave fields. It is most convenient to consider the acoustic pressure wave field $P_j(\boldsymbol{r}, \omega)$; $j = 1, 2$, in the geometry of Fig. 1. With only a change in the transport parameters $(k_j, D_{tj}) \leftrightarrow (c_j, D_j)$, $j = 1, 2$, where c_j is the

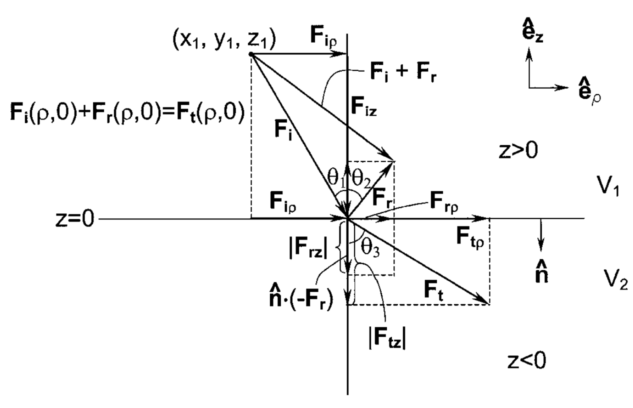

FIG. 2. Interface diffusion-wave flux vector relations under accumulation conditions.

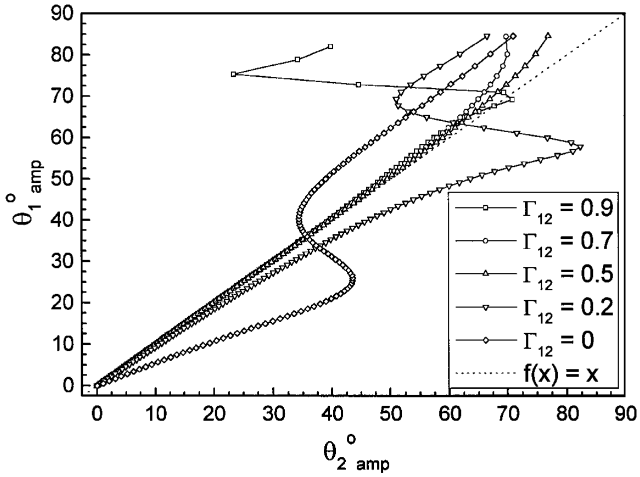

FIG. 3. Relationship between amplitudes of angle of incidence θ_1 and angle of "reflection," θ_2, under accumulation conditions corresponding to Fig. 2. The distance from the point source at $\boldsymbol{r}_1$ was kept constant, $d = \sqrt{\rho^2 + z^2}$.

acoustic velocity and D_j is the density of medium (j), Eq. (1) must be replaced by the Helmholtz equation $\nabla^2 P_j(\boldsymbol{r}, \omega) + k_j^2(\boldsymbol{r}, \omega) P_j(\boldsymbol{r}, \omega) = Q(\boldsymbol{r}, \omega)$; $k_j = \omega / c_j$. The measured acoustic signal is proportional to the pressure intensity vectors $\boldsymbol{I}_j(\rho, z; \omega) = -\frac{i}{2\omega D_j} \nabla(|P_j(\rho, z; \omega)|^2)$, which are gradients of square-law fields [7]. These are directional with angles θ_1 (incident), θ_2 (reflected), and θ_3 (refracted) as conventionally shown in an interface acoustic "ray" diagram. An intensity conservation principle at the interface may be derived if one is careful to convert the non-normal transmitted intensity to total acoustic power transport per unit area of the surface as follows: $\boldsymbol{I}_t^*(\rho, 0; \omega) = (\cos\theta_3 / \cos\theta_1) \boldsymbol{I}_t(\cos, 0; \omega)$. Then it is straightforward to show that the norm of the incident intensity is equal to the sum of reflected plus transmitted intensity norms: $\|\boldsymbol{I}_i(r, 0; \omega)\| = \|\boldsymbol{I}_r(\rho, 0; \omega)\| + \|\boldsymbol{I}_t^*(\rho, 0; \omega)\|$. This is the conventional scalar wave-field conservation law for all values of the interface acoustic coupling coefficient G_{12}. It is mathematically similar to the diffusion-wave flux condition under accumulation, but not under depletion. Furthermore, the reflection law $|\theta_1| = \theta_2$ and Snell's law $(\sin\theta_1 / \sin\theta_3) = c_1 / c_2$ are readily obtained from $\boldsymbol{I}_j(\rho, z; \omega) = -\frac{i}{2\omega D_j} \nabla |P_j(\rho, z; \omega)|^2$. For parabolic diffusion-wave fields the square-law reflection-refraction principles must be replaced by field-gradient-driven accumulation-depletion rules consistent with unidirectionality. These interfacial phenomena can account for the experimentally observed distortions of equisignal contours [4] without the invocation of "internal reflections, diffraction, aberrations, and spurious boundary effects." They tend to limit the accuracy and resolution of imaging techniques such as diffuse photon density waves to angles no larger than 30°–40° from the normal, because the very-near-field nature of the DWF critically depends on interfacial flux processes. In contrast, energy propagation of well-known hyperbolic wave fields, including spatially damped oscillations, is detected with instruments that are sensitive to the amplitude squared, such as electromagnetic power decay within a skin depth in good conductors and eddy currents. These fields exhibit multidirectional properties, including their ability to be "beamed" (guided directionality), and conventional reflection and refraction behavior at interfaces. In conclusion, the historical and widespread interpretation of DWFs in terms of propagating hyperbolic wave-field concepts at interfaces, including diffusion-limited multiply scattered waves [2–4], is fundamentally inconsistent with their parabolic and Fickian nature.

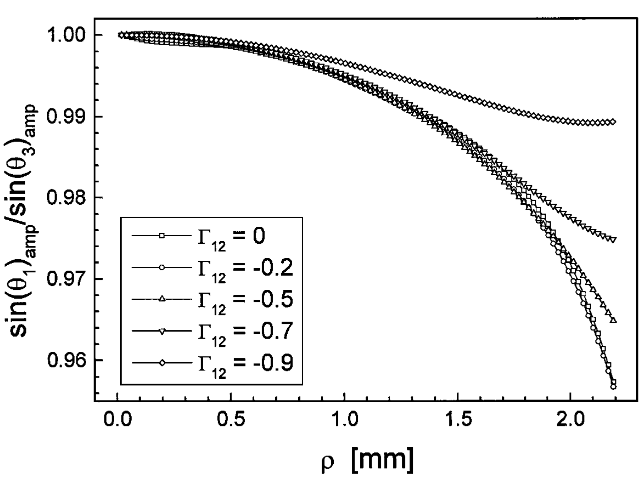

FIG. 4. "Snell's law" formulation under depletion conditions. The distance from the point source at $\boldsymbol{r}_1$ was kept constant, $d = \sqrt{\rho^2 + z^2}$.

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