One-to-One Correspondence between Slowly Decaying Interfacial Profiles and Reflectivity

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A variety of mechanisms give rise to interfacial profiles which approach their bulk values asymptotically like $az^{-\mu}$; for most of them $0<\mu\leqq 2$. In these cases the exponent as well as the amplitude can be determined uniquely by reflectivity measurements slightly above the angle of total external reflection.

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The width of the transition region between two coexisting phases of matter is roughly given by the correlation length ξ for the order parameter (OP) Φ, which distinguishes between these phases. Typically, outside this inhomogeneity, $\Phi(z)$ approaches its bulk value $\Phi(\infty)$ *exponentially*, where z measures the distance from the position of the planar mean interface. However, long-range interactions, the appearance of massless Goldstone modes, or strong collective phenomena can lead to a slow decay according to a *power law* $\Phi(z)-\Phi(\infty)\sim az^{-\mu}$ for $z\rightarrow\infty$. This behavior is encountered in a variety of systems and has attracted substantial theoretical interest. The van der Waals interaction leads to $\mu=3$ for fluid interfaces at low temperatures T.[1] Because of spin waves, $\mu=1$ for the profile of the spontaneous magnetization in a semi-infinite XY or Heisenberg ferromagnet.[2] $\mu=\beta/\nu$ for critical adsorption in one-component fluids or binary liquid mixtures at a wall as well as for complete interfacial wetting at a critical end point of binary liquid mixtures.[3] Here and below, β and ν are the standard critical bulk exponents associated with the actual type of phase transition. For the above examples $\mu=0.5160\pm0.0035$.[4] The same value of μ is obtained for a semi-infinite Ising ferromagnet at criticality, i.e., $T=T_c$, exposed to a surface magnetic field, whereas for an XY and Heisenberg ferromagnet $\mu=0.5165\pm0.0045$ and $\mu=0.5170\pm0.0055$, respectively.[5] For critical adsorption of a tricritical fluid $\mu=\frac{1}{2}$.[6] The monomer concentrations in dilute and semidilute polymer solutions near a wall also exhibit a power-law decay.[7] For both types of solutions, $\mu=3-\nu^{-1}=1.2995\pm0.0040$ below, and $\mu=1-(1-\phi_s)\nu^{-1}$ at the adsorption temperature. Whereas in the previous examples μ is either a rational number of given by *bulk* critical exponents, in the latter case μ is determined by the critical *surface* exponent ϕ_s. With various methods[8] one obtained $0.59\lesssim\phi_s\lesssim0.67$, so that $\mu=0.37\pm0.07$.

These power laws are intimately related to our present understanding of inhomogeneous systems. Therefore, experimental tests for these power laws are particularly interesting. Indeed, numerous experiments have been performed with use of a variety of techniques. All of them, however, suffer from severe limitations with respect to a crucial test of the theoretical predictions. Gravimetric and volumetric experiments[9] are only global measurements. Fluorescence induced by an evanescent wave[10] does provide a spatial resolution, but the exciting wave is a complicated functional of the unknown profile. Furthermore, an absolute measurement of the amplitude a requires detailed knowledge about the fluorescence process. Up to now it was not possible to deconvolute ellipsometric[11] and reflectivity[12] data in order to obtain $\Phi(z)$. At the best one can check the consistency of the data with the numerical solution of Maxwell's equations for a profile one has guessed. One can try to achieve an approximate deconvolution by application of the first Born approximation.[13] But for a pure algebraic decay of $\Phi(z)$ the Born expressions are unreliable, because for $\mu\leqq1$ they predict a divergence of the reflectivity at the angle of total reflection. Yet even in the case of faster-decaying profiles, multiple scattering cannot be neglected for the specular beam so that the Born approximation is inadequate. Solutions based on variational trial functions[14] represent improvements but they show the same kind of divergence. Other inversion methods[15] require a strictly finite interfacial region, which is not the case here.

In this paper we present exact and analytic results for the reflectivity, which allow one to extract uniquely the asymptotic behavior of $\Phi(z)$ from experimental data. A detailed numerical investigation shows that such experiments seem to be feasible.

The propagation of neutrons and electromagnetic waves in the interfacial region is governed by the one-dimensional wave equation

$$[d^2/dz^2+E-V(z)]\psi(z)=0. \tag{1}$$

To be specific, we consider neutrons probing the critical adsorption of a one-component fluid f at a wall w. f (w) fills the half-space $z>0$ ($z<0$). The neutrons of mass m_n, which are incident from the left with energy $\tilde{E}$, experience a potential energy $\tilde{V}(z>0)=2\pi\hbar^2m_n^{-1}b_fc(z)$ with $\tilde{V}_b=\tilde{V}(\infty)$ and $\tilde{V}(z<0)=\tilde{V}_i=2\pi\hbar^2m_n^{-1}\sum_j b_w^{(j)}\times c_w^{(j)}$. b_f and $b_w^{(j)}$ are the scattering lengths of the fluid particles with number density $c(z)$ and of the wall particles with number densities $c_w^{(j)}$, respectively. α denotes the angle between the incident beam and the plane $z=0$.

The neutron wave function

$$\tilde{\psi}(\mathbf{r}=(\mathbf{r}_{\parallel},z))=\psi(z)\exp(i\mathbf{k}_{\parallel}\cdot\mathbf{r}_{\parallel})$$

is determined by Schrödinger's equation from which one obtains Eq. (1) with $V(z)=2m_n\hbar^{-2}[\tilde{V}(z)-\tilde{V}_b]$ and

$$E=2m_n\hbar^{-2}(\tilde{E}-\tilde{V}_i)(\sin^2\alpha-\sin^2\alpha_c).$$

$k_{\parallel}=\hbar^{-1}[2m_n(\tilde{E}-\tilde{V}_i)]^{1/2}\cos\alpha$ is the length of the parallel wave vector. We assume $\tilde{V}_b>\tilde{V}_i$ so that total external reflection occurs for

$$\alpha<\alpha_c=\arcsin[(\tilde{V}_b-\tilde{V}_i)/(\tilde{E}-\tilde{V}_i)]^{1/2}.$$

For $\alpha\rightarrow\alpha_c$, E vanishes like $m_n\hbar^{-2}(\tilde{E}-\tilde{V}_i)\alpha_c(\alpha-\alpha_c)$. In the present case the OP is $\Phi(z)=c(z)-c_c$, where c_c is the number density of the fluid at $T=T_c$. [$\Phi(\infty)$ vanishes at T_c.] This leads to $V(z>0)=4\pi b_f[\Phi(z)-\Phi(\infty)]$. V_i denotes $V(z<0)$. Similar relations are valid for x rays and light. These formulas as well as the applications to the variety of phenomena mentioned at the beginning cannot be presented here for reasons of space.[16] In the following we consider potentials with the asymptotic behavior $V(z\rightarrow\infty)=\lambda z^{-\mu}$. In each case, λ is a known function of the coefficient a; in the above example $\lambda=4\pi b_f a$.

The solutions of Eq. (1) have the form $\psi(z<0)=Ae^{ik_iz}+Be^{-ik_iz}$ with $k_i=(E-V_i)^{1/2}$ and $\psi(z\rightarrow\infty)=C\exp[iE^{1/2}zg(z)]$. $|A|^2$ is the intensity of the incident beam. B and C are determined by the continuity of $\psi(z)$ and $\psi'(z)$ at $z=0$. For $\mu>1$, $g(z)=1$, but for $\mu\leqq 1$, $\psi(z\rightarrow\infty)$ is no longer a plane wave; e.g., for $\frac{1}{2}<\mu<1$, $g(z)=1-\lambda z^{-\mu}/[2(1-\mu)E]$. The reflectivity coefficient can be expressed as

$$R=|[k_i\psi(0)+i\psi'(0)]/[k_i\psi(0)-i\psi'(0)]|^2.$$

We have $R=1$ for $V_i<E<0$ and $R<1$ for $E>0$.

Our first important observation is that the type of nonanalyticity of $R(E)$ at the threshold $E=0$, i.e., at the angle of total reflection $\alpha=\alpha_c$, depends only on the asymptotic behavior of $V(z)$ for $z\rightarrow\infty$. Consider two arbitrary potentials $V_1(z)$ and $V_2(z)$ which differ within the finite interval $z\in(0,z_1)$ but coincide outside this interval, i.e., in particular they have the same asymptotic behavior $V(z\rightarrow\infty)$. The two reflectivity coefficients R_1 and R_2 belonging to V_1 and V_2, respectively, have the following property: With

$$\lim_{E\rightarrow 0^+}\{[1-R_1(E)]/f(E)\}=r_1=\text{const}\in\mathbb{R}^+$$

one can find another positive constant $r_2\in\mathbb{R}^+$ such that for the *same* function

$$f(E)\lim_{E\rightarrow 0^+}\{[1-R_2(E)]/f(E)-r_2\}=0.$$

The proof of this theorem[16] is based on the fact that Eq. (1) allows one to express $\psi(0)$ and $\psi'(0)$ as linear combinations of $\psi(z_1)$ and $\psi'(z_1)$.

In our cases of potentials decaying as $V(z\rightarrow\infty)=\lambda z^{-\mu}$ this theorem states that close to the angle of total reflection the reflectivity coefficient depends—up to an amplitude—only on λ and μ. The universal function $f(E)$ shows the following remarkable dependence on μ (for $E\rightarrow 0$):

$$f(E)=E^{1/2}\text{ for }\mu\in(2,\infty], \tag{2}$$

$$f(E)=E^{(\lambda+1/4)^{1/2}}\text{ for }\mu=2, \tag{3}$$

$$f(E)=\exp[-2I(\mu)\lambda^{1/\mu}/E^{1/\mu-1/2}]\text{ for }\mu\in(0,2). \tag{4}$$

Equation (3) is valid for $\lambda>-\frac{1}{4}$, whereas Eq. (4) holds for $\lambda>0$. For negative values of λ, R can diplay a rather exotic behavior for $E\rightarrow 0$. For example, in the case $\mu=2$, $\lambda<-\frac{1}{4}$, $\lim_{E\rightarrow 0^+}R(E)$ does not exist, but $E=0$ is an accumulation point of resonances with $\sup_E\{R(E)\}<1$.[16]

All potentials which decay faster than z^{-2}, i.e., including an exponential decay, yield at the critical angle a *square-root singularity* of R with a positive curvature. $\mu=2$ is a marginal case, in which one finds a *nonuniversal singularity:* The exponent becomes a continuous function of the strength λ of the potential [Eq. (3)]. For weak potentials, i.e., $\lambda\rightarrow 0$, one recovers the square-root singularity for fast-decaying profiles. For stronger potentials the curvature of $R(E=0^+)$ switches from positive values for $\lambda<\frac{3}{4}$ to negative values for $\lambda>\frac{3}{4}$. For $\lambda\rightarrow\infty$ the singularity becomes of infinite order. This matches smoothly with the *essential singularity* found for $0<\mu<2$. The function $I(\mu)$ in Eq. (4) is universal:

$$I(\mu)=\sum_{n=0}^{\infty}c_n\{[1+\mu(n-\tfrac{1}{2})]n!\}^{-1}, \tag{5}$$

with $c_n=-\Gamma(n-\frac{1}{2})/(2\pi^{1/2})$. $I(\mu)>0$ diverges for $\mu\rightarrow 2$ as $(1-\mu/2)^{-1}$; $I(\mu\rightarrow 0)=(\pi^{1/2}/2)\mu^{1/2}$ (see Fig. 1).

Equation (2) is verified numerically and agrees with the exact solutions for the square-well potential, i.e.,

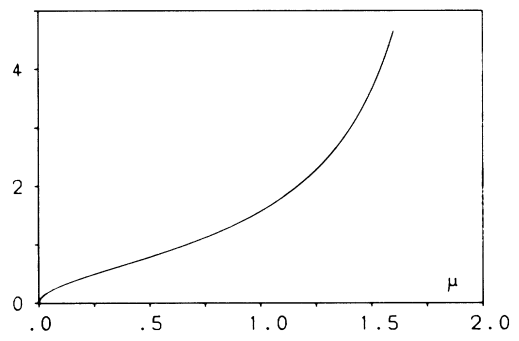

FIG. 1. The universal function $I(\mu)$ characterizes the reflectivity of interfacial profiles decaying as $z^{-\mu}$ with $\mu<2$ [see Eqs. (4) and (5)].

$V(z>z_1)=0$ and $V(0<z<z_1)=\text{const}$, and for $V(z)=\lambda\exp(-z/\chi)$ as well as other exponentially decaying potentials.[16,17a] Equation (3) follows from the exact solution for $V(z)=\lambda(z+z_0)^{-2}$.[16,17] [According to the above theorem the resulting function $f(E)$ is independent of z_0, which checks with the explicit solution.] Equations (4) and (5) are obtained by use of the WKB approximation for $V(z)=\lambda(z+z_0)^{-\mu}$ with $\lambda>0$.[16,18] Although this is an approximation, our numerical analysis[16] yields very strong evidence that Eqs. (4) and (5) are *exact*. This may be understood in terms of the criterion $\gamma=|[E-V(z)]^{-3/2}V'(z)|/2\ll 1$ for the validity of the WKB approximation.[18] The above potentials lead to $\gamma(E\to 0)=\mu\lambda^{-1/2}(z+z_0)^{-1+\mu/2}/2$. In those cases where $\mu<2$ γ becomes arbitrarily small for $z\to\infty$. Together with the above theorem it is therefore no surprise that the WKB expression predicts the correct E dependence of $R(E\to 0,\mu<2)$. This argument is confirmed by the agreement of Eqs. (4) and (5) with the exact result for $\mu=1$.[16,19]

With the exception of the van der Waals tails ($\mu=3$) all other known examples with a power-law decay of the OP profile have $\mu<2$. To determine μ one therefore has to plot the reflectivity data $y=\ln[1-R(E)]$ as a function of $x=E^{1/2-1/\mu'}$. Then the actual exponent μ follows a that value $\mu'=\mu$ which renders a linear function $y(x)$ for $x\to\infty$. The slope y/x of this straight line yields the strength of the potential: $\lambda=|(y/x)/[2I(\mu)]|^{\mu}$. This allows an absolute measurement of the amplitude a of the OP profile, because $\lambda(a)$ (see above) is a known function which depends on the scattering process and the nature of the OP.

μ and λ can only be determined in this way if the asymptotic behavior given in Eq. (4) is experimentally accessible. The answer to this question depends on the system under consideration. In the following we confine ourselves to the much studied case of critical adsorption. There are several facts which limit the observation of Eq. (4) to a window $E_-\lesssim E\lesssim E_+$. Of course, even for a pure power-law potential, the solution of Eq. (1) yields an $R(E)$ which deviates from the asymptotic behavior [Eq. (4)] for larger values of E. With a prescribed accuracy $|y(x)-y_{\text{asymp}}(x)|\leqq\varepsilon$ this leads to an upper bound $E_{\ddagger}^{(1)}$. Even at T_c the OP profile, and therewith $V(z)$, does not decay with a single power law. There are corrections to the leading behavior of the form[3] $\Phi(z)=az^{-\beta/\nu}(1+dz^{-1}+\ldots)$. These corrections become visible for higher values of E and lead to another upper bound $E_{\ddagger}^{(2)}$, so that $E_+=\min(E_{\ddagger}^{(1)},E_{\ddagger}^{(2)})$. In practice, the finite temperature resolution leads to $\xi=\xi_0^+t^{-\nu}\lesssim\xi_{\max}<\infty$, with $t=(T-T_c)/T_c$. As a consequence, $\Phi(z)-\Phi(\infty)\sim\exp(-z/\xi)$ for $z\gtrsim\xi$ and thus is no longer $\sim z^{-\beta/\nu}$. For $\xi\to\infty$, i.e., $T\to T_c$, the exponential decay disappears and one is left with the power law. Because the exponential decay of $V(z)$ leads to a square-root singularity of $R(E\to 0)$, at $E=E_-$ there is a crossover of $R(E)$ from Eq. (4) for $E>E_-$ to Eq. (2) for $E<E_-$. Furthermore, the absorption of the incoming beam leads to $R(E<0)<1$ so that the singularity of R at $E=0$ is smeared out. Therefore Eq. (4) prevails only if the absorption is sufficiently small. The influence of these various contributions must be checked numerically. For that purpose we chose the following model potential:

$$V(z)=\lambda(\mu\xi)^{-\mu}\{\sinh[(z+z_0)/(\mu\xi)]\}^{-\mu}+iV_a. \quad (6)$$

$V_a=\text{Im}V(z)<0$ takes into account the absorption. The choice of Eq. (6) relies on several merits. With $\mu=\beta/\nu$, ReV obeys the scaling form $\sim t^{\beta}F(z/\xi)$ as theoretically expected[3]; there is a smooth crossover from the behavior $\sim z^{-\beta/\nu}$ for $z_0\lesssim z\lesssim\xi$ to $\sim\exp(-z/\xi)$ for $z\gtrsim\xi$, and the sinh term corresponds to the mean-field shape function of the OP. As an example we studied neutron scattering of SF_6 adsorbed at a wall,[9b] i.e., $c_c\cong 3.0\times 10^{21}\,\text{cm}^{-3}$ and $b_f\cong 3.6\times 10^{-12}\,\text{cm}$ so that $m_n\hbar^{-2}\tilde{V}_b=1.4\times 10^{-5}\,\text{Å}^{-2}$. The effect of the OP profile is enhanced if $\tilde{V}_b-\tilde{V}_i$ is small. On the other hand, this difference should be not too small in order to have a reasonable value of α_c. Given the broad spectrum of values for $b_w^{(j)}$ it seems not to be very difficult to find such a suitable wall material. We chose $2m_n\hbar^{-2}\tilde{V}_i=1.2\times 10^{-5}\,\text{Å}^{-2}$ so that $V_i=-2\times 10^{-6}\,\text{Å}^{-2}$. The extinction in SF_6 is dominated by the absorption cross section of the sulfur atoms; the contributions due to incoherent scattering and critical turbidity are negligibly small. For neutrons with a vacuum wavelength $\Lambda\gtrsim 1$ Å, $|V_a|=10^{-10}\,\text{Å}^{-2}$ is an upper bound.[20] In Eq. (6) we chose $\mu=0.5$, $\xi=6000$ Å, and $z_0=10$ Å which corresponds to the choice $d=-5$ Å. The conservative assumption $c(0)=1.5c_c$ [compared with $c(0)\gtrsim 2c_c$ as estimated in Ref. 9b)] leads to $\lambda=2.2\times 10^{-5}\,\text{Å}^{-1.5}$. If the reflectivity R, as calculated numerically from Eq. (6) with the above choice of parameters, is plotted as $y=\ln(1-R)$ vs $x=E^{-1.5}$, which corresponds to our input value $\mu=0.5$, then in the interval

$$E_-=0.95\times 10^{-6}\,\text{Å}^{-2}\leqq E\leqq E_+=4.22\times 10^{-6}\,\text{Å}^{-2}$$

one ends up with a curve, which deviates from a straight line less than $\varepsilon=0.01$, i.e., less than about 1%. From this we conclude that even in the presence of the above-mentioned disturbing effects occurring in reality, the correct exponent of the underlying power law of the OP can be recovered within the window

$$\alpha_-=0.278\times\Lambda\,\text{mrad}\,\text{Å}^{-1}\leqq\alpha\leqq\alpha_+$$
$$=0.461\times\Lambda\,\text{mrad}\,\text{Å}^{-1}.$$

The angle of total reflection is $\alpha_c=0.225\times\Lambda\,\text{mrad}\,\text{Å}^{-1}$. For cold neutrons with $\Lambda=20$ Å this leads to $\alpha_c=4.5$ mrad, $\alpha_-=5.6$ mrad, and $\alpha_+=9.2$ mrad. These limits are accessible with present techniques.[21] We want to emphasize that the only purpose of the above example is

to demonstrate that the data inversion can be accomplished under realistic conditions. Neutron scattering can be applied to all the other examples with a slowly decaying OP. Neutrons are favorable because of their small extinction and because, e.g., deuteration[22] allows one to tailor the necessary condition $\lambda > 0$. Our results apply equally well to the reflectivity of x rays. There the extinction is larger but can be suppressed by use of hard x rays. This lowers the value of α_c but it might be countered by the much higher resolution which can be achieved with synchroton sources. In general we expect that one can achieve a very high resolution, because one is operating right at the maximum of the intensity ($R \approx 1$). For x rays one must use a system in which the electron density in the bulk ($z = \infty$) is higher than in the wall medium ($z < 0$). For example, the wetting of a light wall or of vapor by the heavier liquid phase of a binary liquid mixture corresponds to such a situation. In each case the feasibility can easily be tested in advance by the solution of Eqs. (1) and (6) numerically. Finally we note that Eq. (4) is inaccessible by light scattering, because since $\Lambda/\xi \gtrsim 1$ one has not sufficient spatial resolution. For x rays and neutrons $\Lambda/\xi \lesssim 3 \times 10^{-3}$.

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