Reply to "Comment on 'Fluctuating interfaces in microemulsion and sponge phases'"

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(Received 31 January 1995)

In reply to the preceding Comment of Pieruschka, Safran, and Marčelja, we show that the scattering intensity of level surfaces of Gaussian random fields has the same asymptotic behavior for small wave vectors, both for interfaces of constant and of fluctuating thickness. In the former case, Monte Carlo simulation results for the film correlation function in a Ginzburg-Landau model are found to be in very good agreement with variational results; in particular, the oscillatory component of the correlation function is found to be much more pronounced than for interfaces of fluctuating thickness.

PACS number(s): 82.70.−y, 05.40.+j, 61.20.Gy

In their Comment [1], Pieruschka, Safran, and Marčelja argue correctly that the scattering intensity of a film of (nearly) constant thickness ϵ (with ϵ sufficiently small) is given by

$$G_{\rm film}^{\rm (const)}(\mathbf{r}) \sim \text{Prob}\left[|\Phi(\mathbf{r})| \le \frac{\epsilon}{2}|\nabla\Phi(\mathbf{r})|,\ |\Phi(\mathbf{0})| \le \frac{\epsilon}{2}|\nabla\Phi(\mathbf{0})|\right] , \tag{1}$$

whereas the expression

$$G_{\rm film}^{\rm (fluct)}(\mathbf{r}) \sim \langle \delta_\epsilon(\Phi(\mathbf{r}))\ \delta_\epsilon(\Phi(\mathbf{r}))\rangle , \tag{2}$$

with $\delta_\epsilon(\Phi) = 1/\epsilon$ for $|\Phi| \le \epsilon/2$ and zero otherwise, which we have used in Ref. [2], describes a film of fluctuating thickness. In the limit $\epsilon \to 0$, Eq. (1) can be written as [1]

$$G_{\rm film}^{\rm (const)}(\mathbf{r}) \sim \langle |\nabla\Phi(\mathbf{r})|\ \delta(\Phi(\mathbf{r}))\ |\nabla\Phi(\mathbf{0})|\ \delta(\Phi(\mathbf{r}))\rangle , \tag{3}$$

which clearly shows the additional "measure" factors compared to (2).

Pieruschka and Marčelja already pointed out in Ref. [3] that two level cuts of a three-dimensional field create a film of nonuniform thickness, but also note that Eq. (2) is an approximation to the correct expression, which should be valid for not too small values of r. With the analytic results for the asymptotic behavior of both Eq. (2) (see Refs. [3,2]) and Eq. (3) (see the preceding Comment), this assumption can now be checked explicitly. For films of fluctuating thickness, one finds

$$G_{\rm film}^{\rm (fluct)}(\mathbf{r}) \sim \left[1 - g(r)^2\right]^{-1/2} \tag{4}$$

in the limit $\epsilon \to 0$, where $g(r) = \exp(-r/\xi)\sin(k_0 r)/(k_0 r)$. Thus, the leading spatial dependence of $G_{\rm film}^{\rm (fluct)}(\mathbf{r})$ for large r is given by $\exp(-2r/\xi)[a_0 - a_1\cos(2k_0 r + \gamma)]/(k_0 r)^2$, where $a_0 = a_1 = 1$ and $\gamma = 0$. The same asymptotic form has indeed been found in Ref. [1] for films of constant thickness, although the constants a_0, a_1, and γ are different. The Fourier transform of the term proportional to a_0 dominates the scattering intensity in the regime of small wave vectors k. Therefore, the low-k behavior in *both* cases is given by

$$G_{\rm film}(k) \sim \text{const} + \frac{1}{k}\arctan\frac{k\xi}{2} , \tag{5}$$

in agreement with the results of Ref. [4]. However, the

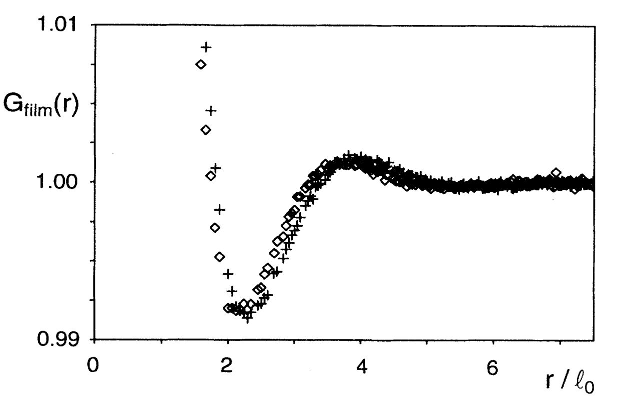

FIG. 1. Normalized correlation function $G_{\rm film}^{\rm (const)}(\mathbf{r})$ for films of (nearly) constant thickness $\epsilon = 0.1\ell_0$. The crosses (+) show the simulation results for level surfaces of Gaussian random fields, the diamonds ($\diamond$) for level surfaces of a Ginzburg-Landau model (compare Ref. [2]). The simulations are carried out on a simple cubic $N \times N \times N$ lattice with $N = 27$ and lattice constant $a_0 = 0.5\ell_0$; averages are taken over about 2×10^6 Monte Carlo steps per site. The parameters of the Ginzburg-Landau model are $g_0 = -2.2$ and $f_0 = 0.5$. The parameters of the Gaussian model are obtained by a variational minimization procedure, as described in Ref. [2].

film correlation function also has an oscillatory component; if $|a_1/a_0|$ is sufficiently large, these oscillations can give rise to a peak in the scattering intensity at $k \simeq 2k_0$ [5] [which is not the case for Eq. (4)].

For films of fluctuating thickness, the data presented in Fig. 9 of Ref. [2] are consistent with the asymptotic behavior (5). We want to mention parenthetically that the dependence of the correlation function (2) on ϵ is rather weak for $\epsilon \ll \langle \Phi^2 \rangle^{1/2}$, so that (5) applies for small ϵ also.

We have used a variational approach in Ref. [2] in order to calculate the film scattering intensity for a Ginzburg-Landau model [6,7] of microemulsion and sponge phases [8]. For films of fluctuating thickness, it was found that the variational results failed to reproduce a peak in the scattering intensity, when they were compared with Monte Carlo data for the Ginzburg-Landau model [2]. We have thus repeated our simulations for the correlation function

$$G_{\text{film}}^{(\text{const})}(\mathbf{r}) \sim \langle |\nabla\Phi(\mathbf{r})| \; \delta_\epsilon(\Phi(\mathbf{r})) |\nabla\Phi(\mathbf{0})| \; \delta_\epsilon(\Phi(\mathbf{r})) \rangle \; , \quad (6)$$

of films of (nearly) constant thickness. Equation (6) is a somewhat different approximation than Eq. (1) to describe films of finite thickness; we prefer (6) since it is easier to handle numerically. Obviously, the expression (3) is recovered in the limit $\epsilon \to 0$.

The correlation function (6) is calculated in both models by Monte Carlo simulations. The results are shown in Fig. 1 — normalized in both cases such that $\lim_{r\to\infty} G_{\text{film}}(\mathbf{r}) = 1$. We find that the oscillations of the variational correlation function are much more pronounced than in the case (2). The agreement of the variational approach with the full Ginzburg-Landau model is remarkable. From this result, together with other results presented in Ref. [2], we can draw the conclusion that in balanced microemulsions level surfaces of Gaussian random fields describe the geometry and structure of the amphiphile film in a Ginzburg-Landau model very well. On the other hand, the rather pronounced disagreement in the case of Eq. (2) indicates that $\nabla\Phi$ at the level surface must vary much more strongly in Gaussian random fields than in a Ginzburg-Landau model. This is not surprising, because the Ginzburg-Landau model contains stable interfaces between an oil-rich and a water-rich phase, while Gaussian random fields do not have any "real" interfaces. This also explains why a peak at $k \simeq 2k_0$ appeared in the film scattering intensity of the Ginzburg-Landau model even with expression (2), while such a peak was absent in variational results [2]. Finally, note that in the limit of strongly swollen microemulsion or sponge phases, where the domain size k_0^{-1} is much larger than the interface width, the expressions (2) and (3) become identical in a Ginzburg-Landau theory.

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[8] The variational approach has been applied to microemulsions by P. Pieruschka and S.A. Safran [Europhys. Lett. **22**, 625 (1993)], who used a curvature model to calculate the scattering intensity in bulk contrast.