Weichman and Goodstein Reply: Phillips and Larese (PL) in their preceding comment [1] and in their earlier paper (PZL) [2] propose a general explanation of the reentrant layering phenomenon seen in multilayer films of argon on graphite. Based on Monte Carlo simulations of the thermodynamics and correlations of films in the neighborhood of three layers thick, they propose a scenario in which subsurface solidification induced by hydrostatic leads to the half-layer offset in the intermediate temperature reentrant first order layering lines. Since their scenario involves both liquid and solid phases in the film they question our use [3] of the restricted solid-on-solid (RSOS) lattice model, which does not distinguish between liquid and solid condensed phases. As we argue below, the behavior of the third layer is rather different from that of higher layers, where our disordered flat (DOF) phase interpretation [3] is claimed to be valid, and is therefore not a good basis for generalization.

To understand the difference between our point of view and that of PL, it is crucial to consider the full experimental phase diagram of the argon-graphite system, shown in Fig. 1 of our paper [3]. Although the first and second layers of argon have independent two-dimensional solid, liquid, and gas phases, complete with critical points, triple points, and melting transitions, the fourth, fifth, and sixth layers behave rather differently. In particular, they do not have triple points or two-dimensional liquid-gas critical points, but they do have low temperature layering transitions at integer layer coverages, and higher temperature "reentrant" layering transitions at half-integer coverages, "zipped" to the low temperature layering transitions by a zigzagging line of heat capacity peaks. The third layer, on the other hand, is an intermediate case, showing both types of behavior: There is a two-dimensional triple point and a two-dimensional critical point, but there is also the first reentrant layering transition, marked by coexistence between $2\frac{1}{2}$ and $3\frac{1}{2}$ layers, which entrains the melting of the third layer. It is not surprising, therefore, that PZL see evidence of melting associated with that rather complicated situation, but the very different nature of the phase diagram for thicker films makes us skeptical of the generalizations they draw from that observation.

There are also two quantitative reasons for doubting the PZL scenario in third and higher layers. First, if it were due to solidification we would expect the vertical steps in adsorption isotherms that are the signature of the phenomenon to be roughly 10% of a layer in height, the typical density difference between liquids and solids (note, in fact, that for *continuous* two-dimensional melting there is no density difference at all). Instead, all of the data, including PZL's own isotherms, consistently show steps of roughly a full layer. Second, the hydrostatic pressure that is supposed to induce the transition is negligible in the third layer, and smaller yet in higher layers. This point shows up clearly in the energetics: The binding energy of the third layer is little more than $k_BT/10$, and decreases as the cube of the film thickness.

Our own approach was to model the system using the RSOS lattice model with a substrate potential and second neighbor interactions. The apparent absence of melting phenomena leads us to believe that this model provides an adequate description of the thicker films in which the physics approaches that of the bulk interface. The DOF phase predicted by this model then produces the *full step* reentrant layering transitions (coexistence between $n + \frac{1}{2}$ and $n - \frac{1}{2}$ layers). PL are certainly correct in their assertion that our model cannot reproduce *all* of the phenomena in a real film, but they in turn have consistently neglected the thicker film portions of the phase diagram in [3] which we believe contain the essential physics of the reentrant layering phenomenon.

PL also raise the issue of whether or not the fact that the real underlying lattice structure is FCC rather than triangular will affect the predictions of the RSOS model. This certainly should be checked, but all evidence so far indicates that the results are not particularly sensitive to lattice structure. We have considered both square and triangular lattices, while, for example, the original work of Rommelse and den Nijs was based on a bcc lattice [4].

In closing, we should also mention that the DOF phase interpretation also leads to a natural explanation for the previously mentioned zigzag sequence of transitions that "zip" the integer and half integer layering lines together [3]: General theory predicts that the bulk interface preroughening transition can be driven first order in a very natural way, and this leads to the observed behavior in multilayer films [5]. Thus the model has remarkable descriptive powers, and the agreement of its predictions with the experimental data is striking. Its very simplicity, that is shortcoming in thinner films, becomes a virtue in thicker films.

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[1] J. M. Phillips and J. Z. Larese, preceding Comment, Phys. Rev. Lett. **75**, 4330 (1995).
[2] J. M. Phillips, Q. M. Zhang, and J. Z. Larese, Phys. Rev. Lett. **71**, 2971 (1993).
[3] P. B. Weichman, P. Day, and D. Goodstein, Phys. Rev. Lett. **74**, 418 (1995).
[4] K. Rommelse and M. den Nijs, Phys. Rev. Lett. **59**, 2578 (1987).
[5] P. B. Weichman and A. Prasad, California Institute of Technology (to be published).

