Glotzer, Stauffer, and Jan Reply: In the preceding Comment [1], Lefever, Carati, and Hassani claim that our recent Monte Carlo simulations [2] of a phase-separating, "reacting" two-component Ising lattice gas were motivated by a modified Cahn-Hilliard equation [3] that "pushes aside the thermodynamics underlying the reaction-diffusion problem studies," because the reactions and the segregation of the two species were considered to be independent of each other. We strongly disagree with their objections, and take this opportunity to clarify the model. In our simulations, the two species of particles A and B were allowed to undergo a "reaction" from A to B, or vice versa, according to a fixed probability p_R, in addition to the usual Kawasaki exchange dynamics governed by the Boltzmann probability. The reactions were triggered and sustained by an external source (which controls the reaction probability p_R), and were thus, in general, far from equilibrium, and the segregation of the spins—which results upon quenching the system to low temperatures—was assumed to be decoupled from the reaction dynamics. The effect of the reactions on phase separation in critical mixtures was shown to be the nonequilibrium, steady-state stabilization of lamellar patterns.

Coarse graining the phenomenological master equation for this lattice model [4] results in the modified Cahn-Hilliard equation given in Eq. (1) of our paper, and proposed by Glotzer, DiMarzio, and Muthukumar (GDM) [3]. A similar equation was proposed earlier by Huberman for a thermodynamically unstable mixture undergoing autocatalytic reactions far from equilibrium [5]. GDM were motivated by mixtures—such as polymer blends—where far-from-equilibrium reactions between the components of the mixture during phase separation interferes with spinodal decomposition. For example, experiments have shown that laser-controlled cross-linking of blends during phase separation results in fascinating steady-state pattern formation, such as "target" or "bulls-eye," as well as lamellar, patterns [6]. Transesterification of pure A and pure B homopolymers into AB copolymers is another example of a far-from-equilibrium reaction that modifies phase separation [7]. In such systems, the nonequilibrium physics controlling the phase separation of the immiscible components may be assumed to be *independent* of the nonequilibrium physics controlling the reactions. The same approach has been taken by Puri and Frisch in their recent treatment of phase-separating, "reactive" binary mixtures [8], as well as by Huberman.

Because we are concerned with the effect of far-from-equilibrium reactions on spinodal decomposition, we believe the equations of motion describing the nonequilibrium kinetics of the model we have considered do not have to abide by the restrictions suggested by Lefever, Carati, and Hassani. We stress that the steady-state patterns we observe in our simulations are not equilibrium patterns,

but rather steady-state, nonequilibrium patterns sustained by reactions that are maintained far from equilibrium. That is, the patterns arise from a dynamic competition between the phase-separation and the nonequilibrium reaction processes. Thus, a suppression of the growth of long-wavelength concentration fluctuations by the reactions is indeed possible during both the early [3,5] and late [9] stages of spinodal decomposition.

Indeed, chemical reactions in closed, interacting, *equilibrium* mixtures should be treated differently than the systems we considered [10]. However, even there it is not necessarily true that a simple extension of equilibrium chemical thermodynamics in the manner in which Lefever, Carati, and Hassani suggest, will suffice.

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[1] R. Lefever, D. Carati, and N. Hassani, preceding Comment, Phys. Rev. Lett. **75**, 1674 (1995).
[2] S. C. Glotzer, D. Stauffer, and N. Jan, Phys. Rev. Lett. **72**, 4109 (1994).
[3] S. C. Glotzer, E. A. DiMarzio, and M. Muthukumar, Phys. Rev. Lett. **74**, 2034 (1995).
[4] S. C. Glotzer, in *Annual Reviews of Computational Physics,* edited by D. Stauffer (World Scientific, Singapore, 1995), Vol. 2.
[5] B. A. Huberman, J. Chem. Phys. **65**, 2013 (1976).
[6] Q. Tran-Cong *et al.,*, Macromolecules **22**, 2720 (1989); in *Dynamics and Patterns in Complex Fluids,* edited by A. Onuki and K. Kawasaki (Springer-Verlag, New York, 1990), p. 60; Macromolecules **24**, 1505 (1991); Polym. Eng. Sci. **33**, 772 (1993).
[7] H. Yoon and C. C. Han, Polym. Eng. Sci. (to be published).
[8] S. Puri and H. L. Frisch, J. Phys. A **27**, 6027 (1994).
[9] S. C. Glotzer and A. Coniglio, Phys. Rev. E **50**, 4241 (1994).
[10] M. Gitterman, J. Stat. Phys. **58**, 707 (1990), and references therein.