Comment on "Exact Results for the Lower Critical Solution in the Asymmetric Model of an Interacting Binary Mixture"

In a recent Letter, Lin and Taylor (L-T) [1] reported exact results for a square lattice gas model of an interacting binary mixture. In this model, each cell of the lattice can be occupied by a square (particle *A*) or by four triangles (particles *B*). The nearest-neighbor coupling between the *A* particles is ϵ_{AA}, while the coupling between *A* and *B* particles, ϵ_{AB} is introduced for each edge contact between the squares and triangles. This model can be considered as a generalization of the "venerable" decorated lattice model introduced three decades ago by Widom [2]. With an appropriate transformation, the partition function of this model maps onto the partition function of the two-dimensional Ising model.

L-T found that the necessary condition for the occurrence of a lower critical point (LCP) in addition to the upper critical point (UCP) is that not only $\epsilon_{AA} < 0$ and $\epsilon_{AB} < 0$ but also the ratio $s = -\epsilon_{AA}/(2\epsilon_{AB} - \epsilon_{AA})$ must exceed 1. The aim of this Comment is to show that it is possible to obtain LCP for $\epsilon_{AA} < 0$, $\epsilon_{AB} < 0$, and $\epsilon_{AA} \geq 2\epsilon_{AB}$, which implies $s > 1$ and $s < 0$.

All the lines of critical points for $\epsilon_{AA} < 0$ end at the same point. This point corresponds to the critical pressure p_e and critical temperature of the pure *A* system.

In Fig. 1, the dashed line corresponds to the coexistence line of the *A* pure system. The zone between this line and the line of critical points (continuous line) is where the coexistence of the mixture occurs.

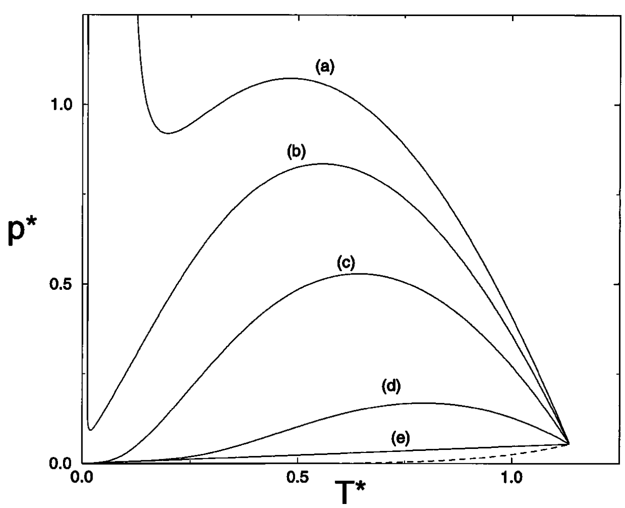

FIG. 1. Lines of critical points (continuous lines) (a) $\epsilon_{AB} = 0.45\epsilon_{AA} (s > 1)$, (b) $\epsilon_{AB} = 0.49\epsilon_{AA} (s > 1)$, (c) $\epsilon_{AB} = 0.6\epsilon_{AA}$, (d) $\epsilon_{AB} = \epsilon_{AA}$, and (e) $\epsilon_{AB} = -\infty$. The reduced units are defined as $p^* = -2p\sigma/\epsilon_{AA}$ and $T^* = -2k_BT/\epsilon_{AA}$ where σ is the area of the lattice cell.

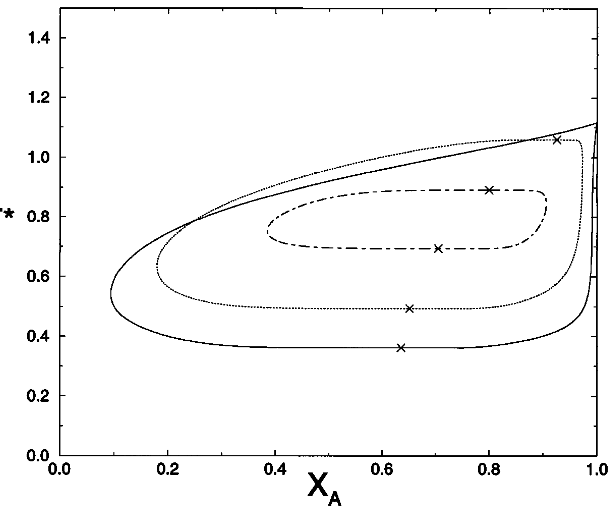

FIG. 2. Coexistence curves at constant pressure for the case (d) of Fig. 1. Continuous line $p < p_e$, dotted line $p_1 > p_e$, and dot-dashed line $p_2 > p_1 > p_e$. The crosses correspond to the location of critical points.

For values of $s < 0$, the line of critical points can present a nonmonotonic behavior with a maximum and then the system exhibits a LCP and an UCP for values of the pressure between this maximum and p_e. For values smaller than p_e, the UCP disappears and the system shows a coexistence with LCP and ending in the pure system (see Fig. 2).

Decreasing ϵ_{AB}, the line of critical points becomes monotonic and consequently, the UCP does not appear and the system shows a single LCP for $p < p_e$. The limit $\epsilon_{AB=-\infty}$ presents a straight line of critical points.

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