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 $e_{AA}$, while the coupling between $A$ and $B$ particles, $e_{AB}$ 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 $e_{AA}, 0$ and $e_{AB}, 0$ but also the ratio $s - 2e_{AA}/2e_{AB} < 0$, which implies $s > 1$ and $s < 0$.

All the lines of critical points for $e_{AA} < 0$ end at the same point. This point corresponds to the critical pressure $p_{cr}$ 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.

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 $p_{cr}$. For values smaller than $p_{cr}$, the UCP disappears and the system shows a coexistence with LCP and ending in the pure system (see Fig. 2).

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

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