Supplemental material to the article

"Lattice Gas Dynamics"

Here, we derive the master equation for dynamic correlation function. Consider three time moments: initial ($t_1 = 0$), intermediate ($t_2 = t$), and final ($t_3 = \Delta t + 1$). The statistical weight of corresponding random process (eq. 11 of main text) is:

$$\begin{aligned}
\Omega\left\{n_1 \to n_2 \to n_3\right\} = \exp\left(\frac{H\left\{n_1(r)\right\} - H\left\{n_3(r)\right\}}{T}\right) \cdot \\
\cdot \int \prod_r db_3(r) db_2(r) dw_3(r) dw_2(r) \cdot \\
\cdot \left(1 + s^-_{2b} b_2\right)\left(1 + s^-_{3b} b_3\right) \cdot \left(1 + s^-_{2w} w_2\right)\left(1 + s^-_{3w} w_3\right) \cdot \\
\cdot \left(1 + \left[s^+_{2b} + (n_1 - s^+_{2b}) b_2\right] \sum_{r'} D_b(r, r', t) b^*_2(r')\right) \cdot \\
\left(1 + \left[s^+_{3b} + (n_2 - s^+_{3b}) b_3\right] \sum_d D_b b^*_3(r + d)\right) \cdot \\
\cdot \left(1 + \left[s^+_{2w} + (1 - n_1 - s^+_{2w}) w_2\right] \sum_{r'} D_w(r, r', t) w^*_2(r')\right) \cdot \\
\cdot \left(1 + \left[s^+_{3w} + (1 - n_2 - s^+_{3w}) w_3\right] \sum_d D_w w^*_3(r + d)\right) \cdot \\
\cdot \exp\left\{-\frac{1}{2}(b_2 b^*_2 + b_3 b^*_3 + w_2 w^*_2 + w_3 w^*_3)\right\}. \qquad (1)
\end{aligned}$$

For arbitrary second time moment $t \neq 1$, one has to treat all spatial lattice sites as nearest neighbors, and use $D_{b,w}(r, r', t)$ instead of diffusion coefficients $D_{w,b}$ (lines 5 and 7 in (1)). To get dynamic correlation function at final moment $t + 1$, let us average (1) over configurations $\{n_1,\ n_2,\ n_3\}$

$$\begin{aligned}
D_{b,w}(r, r', t + 1) = \frac{1}{A} \sum_{\{n_1, n_2, n_3\}}^{\sim} \Omega\left\{n_1 \to n_2 \to n_3\right\}, \\
A = \sum_{\{n_1, n_2, n_3\}} \Omega\left\{n_1 \to n_2 \to n_3\right\}. \qquad (2)
\end{aligned}$$

The "$\sim$" means that the sum covers only those realizations of the process,

which include transition of one and the same black (for $D_b(r,r',t+1)$) or white (for $D_w(r,r',t+1)$) particle from r at $t_1=0$ to r' at $t_3=t+1$. To fulfill this condition, we use known trick which allows to select single polymer line in polymer statistics. Let us introduce into (1) three additional scalar fields. The first of them – y – is complex delta - correlated spatial field. We insert $y(r)$ into every ending of diffusion step of black particle during time $t_1 \to t_2$, and its conjugated value $y^*(r)$ – into every beginning of those during $t_2 \to t_3$. The other two fields ψ_1 and ψ_2 serve as indicators (without integration over them). We put $\psi_1(r)$ into every beginning of black particle diffusion step during $t_1 \to t_2$, and $\psi_2(r)$ – into every ending of those during $t_2 \to t_3$. As a result, one gets the functional of fields $\psi_1,\ \psi_2$

$$\begin{aligned}
\Omega_b\left\{\psi_1,\psi_2\right\} = \sum_{\{n_1,n_2,n_3\}} \exp\left(\frac{H\left\{n_1(r)\right\} - H\left\{n_3(r)\right\}}{T}\right) \cdot \\
\cdot \int \prod_r db_3(r) db_2(r) dw_3(r) dw_2 \cdot \\
\cdot \left(1+s^-_{2b} y b_2\right)\left(1+s^-_{3b} b_3\right)\left(1+s^-_{2w} w_2\right)\left(1+s^-_{3w} w_3\right) \cdot \\
\cdot \left(1+\left[s^+_{2b}+(n_1-s^+_{2b})b_2\right]\psi_1 \sum_{r'} D_b(r,r',t) b^*_2(r') y(r')\right) \cdot \\
\cdot \left(1+\left[s^+_{3b}+(n_2-s^+_{3b})b_3\right] y^* \sum_d D_b b^*_3(r+d)\psi_2(r+d)\right) \cdot \\
\cdot \left(1+\left[s^+_{2w}+(1-n_1-s^+_{2w})w_2\right]\sum_{r'} D_w(r,r',t) w^*_2(r')\right) \cdot \\
\cdot \left(1+\left[s^+_{3w}+(1-n_2-s^+_{3w})w_3\right]\sum_d D_w w^*_3(r+d)\right) \cdot \\
\cdot \exp\left\{-\frac{1}{2}(b_2 b^*_2 + b_3 b^*_3 + w_2 w^*_2 + w_3 w^*_3 + y y^*)\right\}, \qquad (3)
\end{aligned}$$

which is the generating functional for (2):

$$D_b(r,r',t+1) = \frac{1}{A} \cdot \frac{\partial^2 \Omega_b\{\psi_1,\psi_2\}}{\partial\psi_1(r)\ \partial\psi_2(r')}\Bigg|_{\psi_1=\psi_2=1} \qquad (4)$$

The $D_w(r,r',t)$ may be calculated by similar way. The (4), after calculation of functional integrals in (1,3), is the master equation on dynamic correlation function $D_{b,w}(r,r',t)$. The approach may be considered as a lattice gas analogue of the mode coupling theory.

For quasi – binary system, there are no auxiliary field w, i.e. one put $w_2 = w_3 = 0$, and omits integration over these fields. The lower index w is insufficient in this case, so we denote $\Omega_b = \Omega, D_b = D$.

(3) contains filling numbers n_2 as local term, so that sum over these variables may be easily done. To do so over $\{n_1\}, \{n_3\}$, one uses Hubbard – Stratonovich transform to auxiliary fields $x_1(r),\ x_3(r)$. As a result one gets functional integral over x_1, x_3, b_3, b_2, y, with "Lagrangian", containing "free field" quadratic part

$$
\begin{aligned}
&-\frac{T}{2}\sum_{r,r'} x_1(r)J^{-1}(r,r')x_1(r') - \\
&-\frac{T}{2}\sum_{r,r'} x_3(r)J^{-1}(r,r')x_3(r') - \\
&-\frac{1}{2}\sum_{r}(b_2 b_2^* + b_3 b_3^* + yy^*)\ ,
\end{aligned}
\tag{5}
$$

and logarithmic interaction term $\sum_r \ln\Theta(r)$, where

$$
\begin{aligned}
\Theta = 1 + (1-b_2)(1+F) + \\
+e^{\alpha}[1 + (1+F)(1+G)] + \\
+e^{\beta}[1 + b_3 + (1-b_2)(1+b_3F)] + \\
+e^{\alpha+\beta}[1 + b_3 + (1-b_2)(1+b_3F) + \\
+(1+b_3)G + b_2(1+b_3F)(1+G)]\ , \\
F(r) = y^*(r)\sum_{r'} D(r,r')b_3^*(r')\psi_2(r')\ , \\
G(r) = \psi_1(r)\sum_{r'} D(r,r',t)b_2^*(r')y(r')\ .
\end{aligned}
\tag{6}
$$

Expanding the last up to the first order of b_3, b_2, y, and calculating resulting Gauss integrals, one gets

$$
\begin{aligned}
\Omega\{\psi_1,\psi_2\} = \int Dx_1 Dx_3 \exp\left[\Gamma\left\{x_1,x_3,\psi_1,\psi_2\right\}\right], \\
\Gamma\left\{x_1,x_3,\psi_1,\psi_2\right\} = -\frac{T}{2}\sum_{r,r'} x_1(r)J^{-1}(r,r')x_1(r') - \\
-\frac{T}{2}\sum_{r,r'} x_3(r)J^{-1}(r,r')x_3(r') + \ln\left(1+e^{\alpha}\right) +
\end{aligned}
$$

$$+\ln\left(1+e^{\beta}\right)+\frac{1}{128}\sum_{r,r',r''}\left(\frac{e^{\alpha}}{1+e^{\alpha}}\psi_1\right)_{r'}D(r',r,t)\cdot$$
$$\cdot\left(\frac{e^{\alpha}}{(1+e^{\alpha})^2(1+e^{\beta})}\right)_{r}D(r,r'')\left(\frac{e^{\beta}}{1+e^{\beta}}\psi_2\right)_{r''}, \tag{7}$$

where $\alpha = ix_1+\mu_1\,,\quad \beta = x_3+\mu_3$. Chemical potentials $\mu_1(r)$ and μ_3 allow one to specify initial and final sets of configurations, respectively. For integration over x_3, we use the mean field approximation, i.e. maximization of Γ with respect to x_3:

$$\frac{\delta\Gamma\left\{x_1,x_3,\psi_1,\psi_2\right\}}{\delta x_3(r)}=0. \tag{8}$$

Then, one has to average solution of (8) over $x_1(r)$. Instead, we average (8) before its solution:

$$T\sum_{r'}J^{-1}(r-r')x_3(r')=\left(\frac{e^{\beta}}{1+e^{\beta}}\right)_{r}-$$
$$-\frac{1}{128}K(r)\left(\frac{e^{\beta}}{(1+e^{\beta})^2}\right)_{r}\sum_{r'}D(r,r')\left(\frac{e^{\beta}}{1+e^{\beta}}\right)_{r'}+$$
$$+\frac{1}{128}\left(\frac{e^{\beta}}{(1+e^{\beta})^2}\right)_{r}\sum_{r'}D(r,r')K(r')\left(\frac{1}{1+e^{\beta}}\right)_{r'}\,, \tag{9}$$

where

$$K(r)=\sum_{r'}D(r,r',t)\left\langle\left(\frac{e^{\alpha}}{1+e^{\alpha}}\right)_{r}\left(\frac{e^{\alpha}}{(1+e^{\alpha})^2}\right)_{r'}\right\rangle, \tag{10}$$

and angular brackets denote gaussian average

$$\langle..\rangle=\int..Dx_1\exp\left(-\frac{T}{2}\sum_{r,r'}x_1(r)J^{-1}(r,r')x_1(r')\right). \tag{11}$$

Equation (4) takes the form:

$$D(r,r',t+1)=N\sum_{r''}\left\langle\left(\frac{e^{\alpha}}{1+e^{\alpha}}\right)_{r}\left(\frac{e^{\alpha}}{(1+e^{\alpha})^2}\right)_{r''}\right\rangle\cdot$$
$$\cdot D(r,r'',t)D(r'',r')\left(\frac{1}{1+e^{\beta}}\right)_{r''}\left(\frac{e^{\beta}}{1+e^{\beta}}\right)_{r'}. \tag{12}$$

Normalizing multiplier N has to be chosen as to provide

$$\sum_{r'} D(r, r', t) = 1. \tag{13}$$

In (12) we denoted $D(r, r') = D(r, r', t = 1)$. In (13), summation extends over all points, including $r' = r$. Eqs. (9 – 13) provide the necessary master equation. Let us list approximations used:

1. linear expanding of interaction term;
2. mean – field integration over x_3;
3. average of (8) before its solution.