Electronic Supplementary Material (ESI) for Soft Matter

Supplementary information for "Ring formation by competition between entropic effect and thermophoresis"

We derive eq. (8) from a lattice model following ref. [11]. We consider random walk of a tracer (colloid) under the presence of other particles (polymers). Both the tracer and the other particles perform random walks on a d-dimensional regular lattice in which double occupancy of the lattice sites is not allowed. We denote the vector characterizing a jump to the nearest neighbor site j by $\vec{b}_j$ ($j = 1, 2, \cdots, 2d$) and the jump length by b. The probability at time t that the site $\vec{r}$ occupied by a tracer, $c(\vec{r}, t)$, obeys,

$$\frac{\partial}{\partial t} c(\vec{r}, t) = \sum_j \left[\gamma_B \left(\vec{r} + \vec{b}_j \right) P \left(\vec{r} + \vec{b}_j \bullet, \vec{r}\phi, t \right) - \gamma_B \left(\vec{r} \right) P \left(\vec{r}\bullet, \vec{r} + \vec{b}_j \phi, t \right) \right], \qquad \text{(a)}$$

where $\gamma_B \left(\vec{r} \right)$ denotes the transition probability and $P \left(\vec{r}\bullet, \vec{r'}\phi, t \right)$ denotes the joint probability at time t that the site $\vec{r}$ is occupied by a tracer and the site $\vec{r'}$ is empty. $P \left(\vec{r}\bullet, \vec{r'}\phi, t \right)$ can be rewritten as,

$$P \left(\vec{r}\bullet, \vec{r'}\phi, t \right) = c(\vec{r}, t) - P \left(\vec{r}\bullet, \vec{r'}\circ, t \right), \qquad \text{(b)}$$

where $P \left(\vec{r}\bullet, \vec{r'}\circ, t \right)$ denotes the joint probability at time t that the site $\vec{r}$ is occupied by a tracer and the site $\vec{r'}$ is occupied by another particle. By substituting eq. (b), eq. (a) can be expressed as

$$\begin{aligned} \frac{\partial}{\partial t} c \left(\vec{r}, t \right) = \sum_j \Big[\gamma_B \left(\vec{r} + \vec{b}_j \right) \left(c \left(\vec{r} + \vec{b}_j, t \right) - P \left(\vec{r} + \vec{b}_j \bullet, \vec{r}\circ, t \right) \right) - \\ \gamma_B \left(\vec{r} \right) \left(c \left(\vec{r}, t \right) - P \left(\vec{r}\bullet, \vec{r} + \vec{b}_j \circ, t \right) \right) \Big]. \qquad \text{(c)} \end{aligned}$$

We denote the occupation probability by other particles by $p \left(\vec{r}, t \right)$ and consider the case that the joint probability function can be expressed as,

$$P \left(\vec{r}\bullet, \vec{r'}\circ, t \right) = c \left(\vec{r}, t \right) p \left(\vec{r'}, t \right) \sigma(\vec{r} - \vec{r'}), \qquad \text{(d)}$$

where the spatial correlation is taken into account by $\sigma(\vec{r} - \vec{r'})$. By ignoring the spatial variation of $\sigma(\vec{r})$ in the limit of small lattice spacing and introducing definition

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given by $\sigma(0) = \lim_{b\to 0}(\vec{b}_j)$, we obtain,

$$\sum_j \gamma_B\left(\vec{r}+\vec{b}_j\right) P\left(\vec{r}+\vec{b}_j\bullet, \vec{r}\circ, t\right) = \left[\gamma_B\left(\vec{r}\right) c\left(\vec{r},t\right) p\left(\vec{r},t\right) + b^2 p\left(\vec{r},t\right) \nabla^2\gamma_B\left(\vec{r}\right) c\left(\vec{r},t\right)\right] \sigma(0), \tag{e}$$

$$\sum_j P\left(\vec{r}\bullet, \vec{r}+\vec{b}_j\circ, t\right) = \left[c\left(\vec{r},t\right) p\left(\vec{r},t\right) + b^2 c\left(\vec{r},t\right) \nabla^2 p\left(\vec{r},t\right)\right] \sigma(0). \tag{f}$$

When $\gamma_B\left(\vec{r}\right)$ depends on $\vec{r}$ through the spatial variation of temperature, the Soret coefficient can be introduced by

$$S_T^c = \left(\frac{\partial \gamma_B}{\partial T}\right) / \gamma_B. \tag{g}$$

We substitute eqs. (e)-(f) into eq. (c). It is convenient to represent $b^2 p\left(\vec{r},t\right) \nabla^2\gamma_B\left(\vec{r}\right) c\left(\vec{r},t\right)$ in terms of $\vec{\nabla}\cdot b^2 p\left(\vec{r},t\right) \vec{\nabla}\gamma_B\left(\vec{r}\right) c\left(\vec{r},t\right)$ and $b^2 c\left(\vec{r},t\right) \nabla^2 p\left(\vec{r},t\right)$ in terms of $\vec{\nabla}\cdot b^2 c\left(\vec{r},t\right) \vec{\nabla} p\left(\vec{r},t\right)$. By further ignoring the spatial dependence in S_T^c, eq. (c) in the limit of $b \to 0$ becomes,

$$\frac{\partial}{\partial t} c\left(\vec{r},t\right) = \vec{\nabla}\cdot D_B^c\left[(1-\sigma(0)p)\left(c S_T^c \vec{\nabla} T + \vec{\nabla} c\right) + \sigma(0) c \vec{\nabla} p\right], \tag{h}$$

where $D_B^c = b^2\gamma_B$. In the mean field approximation in which $\sigma(0) = 1$, eq. (h) reduces to eq. (8),

$$\frac{\partial}{\partial t} c\left(\vec{r},t\right) = \vec{\nabla}\cdot D_B^c\left[(1-p)\left(c S_T^c \vec{\nabla} T + \vec{\nabla} c\right) + c \vec{\nabla} p\right]. \tag{i}$$

The factor $1-p$ originates from the vacant probability of a neighboring site in the mean-field approximation. $D_B^c(1-p)$ represents the tracer diffusion coefficient.

Finally, we would like to add a remark about one component system, where the jump rate of a tracer particle is the same as that of other particles. If we do not distinguish a tracer from other particles, we can set $c\left(\vec{r},t\right) = p\left(\vec{r},t\right)$. In this case we obtain,

$$\frac{\partial}{\partial t} p\left(\vec{r},t\right) = \vec{\nabla}\cdot D_B^c\left[(1-p) p S_T^c \vec{\nabla} T + \vec{\nabla} p\right], \tag{j}$$

where $p\left(\vec{r},t\right)$ is the concentration of Brownian particles including a tracer. In eq. (j) the diffusion coefficient is given by the collective diffusion coefficient D_B^c. Equation (j) reduces to that in ref. [1] of the Supplementary information.

References

[1] H. Jung, V. E. Gusev, H. Baek, Y. Wang, G. J. Diebold, Physics Letters A **375**, 1917 (2011).