Calculating Propagators and Saving Memory

1. Continuous Gaussian chain

For continuous Gaussian chain used in PSCF, the architecture of an ***acyclic*** block copolymer (BCP) chain, which consists of $n_B \geq 1$ (linear homopolymer) **blocks** connecting $n_V = n_B + 1$ **vertices** (note that an acyclic chain can be constructed by adding, in each step, a block and a new vertex to an existing vertex), including both joints (J) and free ends (E), is described by specifying the vertices j and $k \in [0, n_B]$ connected by each block i (=0,…,n_B−1). Each block i has two (one-end-integrated) propagators $q_i(\mathbf{r},s)$ and $q_i^\dagger(\mathbf{r},s)$, where $\mathbf{r}$ denotes the spatial position, and s=0,…,$N_i\Delta s$ with N_i being the number of steps along the chain contour into which the block is uniformly discretized, $\Delta s \equiv f_i/N_i$ being the step-size, and f_i being the volume fraction of block i in the copolymer chain; in the REPS-K method (K=0,…,4; see REPS.pdf for details), N_i must be an integer multiple of 2^K (*i.e.*, $M_i \equiv N_i/2^K$ must be a positive integer). We calculate in Step I all the forward propagators $q_i(\mathbf{r},s)$, then in Step II all the backward propagators $q_i^\dagger(\mathbf{r},s)$.

Hereafter we take the calculation of propagators for block i as an example. To calculate its contribution to the volume-fraction field (at a spatial position $\mathbf{r}$) of its segment type, we need to evaluate $\int_0^{f_i} \mathrm{d}s q_i(\mathbf{r},s) q_i^\dagger(\mathbf{r},s) \approx \Delta s \sum_{k'=1}^{M_i} \sum_{k=0}^{2^K} c_k q_{i,2^K(k'-1)+k} q^\dagger_{i,2^K(k'-1)+k}$ using the Romberg integration (denoted by RI-K; see RI.pdf for details), where c_k's are the coefficients used in RI-K, $q_{i,j} \equiv q_i(\mathbf{r}, j\Delta s)$, and $q^\dagger_{i,j} \equiv q_i^\dagger(\mathbf{r}, j\Delta s)$. To reduce the memory usage, we use the "slice" algorithm proposed in Ref. 1: We store $q_{i,j}$ for all i at their "**check points**" with j=0 and $j = N_i - \left(\sum_{i=1}^{l} i - 1\right) = N_i - \frac{l(l+1)}{2} + 1$ only for ***positive integer*** values of $l < l_{\max}$ (in the ***descending*** order) in Step I above, where $j > 0 \;\Rightarrow\; l^2 + l - 2(N_i + 1) < 0 \;\Rightarrow\; l_{\max} = \left(\sqrt{8N_i + 9} - 1\right)/2$. In Step II above, we only store $q^\dagger_{i,j^*}$ for the current value of j^* (=N_i,N_i−1,…,0); note that $q^\dagger_{i,j^*-1}$ needs to be calculated out of place in REPS-K due to its successive halving of Δs. If q_{i,j^*} is available (*e.g.*, when j^* is a check point), we then directly calculate the contribution of j^* to the above integral; otherwise, we calculate all $q_{i,j}$ between the corresponding check points enclosing j^* and store them in the same places as the no-longer-needed q-values, then calculate their contribution to the above integral. Compared to the usual approach of calculating and storing all propagators before calculating the volume-fraction fields, the "slice" algorithm reduces the memory usage by a factor of $\sqrt{2N_i}$ for large N_i at the cost of increasing the computation by 50% (*i.e.*, solving $q_{i,j}$ twice).

Similarly, to calculate the contribution of block i to the system stress (which vanishes when the bulk periodicity of an ordered phase formed by BCP self-assembly is found), we evaluate $\sum_{\mathbf{q}} \frac{\mathrm{d}\hat{\Phi}_i(q)}{\mathrm{d}q} \frac{\partial q}{\partial \boldsymbol{\theta}} \int_0^{f_i} \mathrm{d}s \hat{q}_i(\mathbf{q},s) \hat{q}_i^\dagger(-\mathbf{q},s)$ in the same way as above, where $\hat{\Phi}_i(q) = \exp\left(-b_i^2 q^2/6\right)$ with b_i being the statistical segment length of block i, $\hat{q}_i(\mathbf{q},s) \equiv \int \mathrm{d}\mathbf{r} \exp\left(-\sqrt{-1}\mathbf{q}\cdot\mathbf{r}\right) q_i(\mathbf{r},s)/V$ and $\hat{q}_i^\dagger(\mathbf{q},s) \equiv \int \mathrm{d}\mathbf{r} \exp\left(-\sqrt{-1}\mathbf{q}\cdot\mathbf{r}\right) q_i^\dagger(\mathbf{r},s)/V$ with $\mathbf{q}$ being the wavevector and V the system volume,

$q \equiv |\mathbf{q}|$, and the vector $\boldsymbol{\theta}$ denotes the (up to six) unit-cell parameters.

2. Discrete chain

On the other hand, an ***acyclic*** discrete chain used in PSCF+ consists of $n_B \geq 1$ (linear homopolymer) **blocks** connected by $n_J = n_B - 1$ **joint bonds (JBs)** each connecting two end-segments of different blocks (note that an acyclic chain here can be constructed by adding, in each step, a joint bond and a new block to an existing block). Each block i has $N_i \geq 1$ segments, for which two propagators need to be calculated. A block of $N_i = 1$ (*i.e.*, a **joint segment, JS**) is considered as a vertex, and the two end-segments of a block of $N_i \geq 2$ are considered as two vertices connected by a **block bond (BB)** of $N_{i'} \equiv N_i - 2$; a **vertex** can be either a joint (J, not necessarily a JS) or a free end (E, which is an end-segment), and has at most one BB connected to it. The chain then consists of $n_b \geq 1$ **v-bonds**, including both JBs (for which $N_{i'} = 0$) and BBs each characterized by its $N_{i'}$ and effective bond length $b_{i'}$ ($i' = 0,\ldots,n_b-1$), connecting $n_V = n_b + 1$ vertices, and its architecture is described by specifying the vertices j and $k \in [0, n_b]$ connected by each v-bond i'. Each block i (*i.e.*, all vertices and BB i' with $N_{i'} \geq 1$) has two propagators $q_i(\mathbf{r}, s)$ and $q_i^\dagger(\mathbf{r}, s)$, where $s = 1,\ldots,N_i$. We calculate in Step I all the forward propagators $q_i(\mathbf{r}, s)$, then in Step II all the backward propagators $q_i^\dagger(\mathbf{r}, s)$.

Hereafter we take the calculation of propagators for block i as an example. To calculate its contribution to the volume fraction field (at a spatial position $\mathbf{r}$) of the species to which the block belongs, we need to evaluate $\sum_{j=0}^{N_i-1} q_{i,j} q_{i,j}^\dagger$, where $q_{i,j} \equiv q_i(\mathbf{r}, s = j+1)$ and $q_{i,j}^\dagger \equiv q_i^\dagger(\mathbf{r}, s = j+1)$. To reduce the memory usage, the above "slice" algorithm with slight modification will be implemented in PSCF+ soon; same for calculating the contribution of a BCP component due to its bonds to the system stress.

References:

1. Qiang, Y. Chap. 6.3.2 in The High-Performance Algorithms for Self-Consistent Field Theory. PhD Thesis, Fudan University, Shanghai, China, 2022.