Recent developments of molecular dynamics calculation software MODYLAS

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Introduction

Nagoya University (National Univ.)

NU has six Nobel Prize winners:

Chemistry R. Noyori [2001], O. Shimomura [2008]

Physics M. Kobayashi & T. Maskawa [2008], I. Akasaki & H. Amano [2014]

Introduction

Department of Materials Chemistry, Nagoya University

・Theoretical and Computational Chemistry

Other groups:
- Catalyst Design
- Material Design Chemistry
- Energy Conversion Chemistry

Staff of Okazaki Lab.

Professor: Susumu Okazaki

Associate Professor: Wataru Shinoda

Assistant Professor: Kazushi Fujimoto

Designated Associate Prof.: Noriyuki Yoshii (CCS) →Theory of FMM

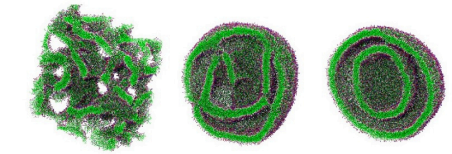
Designated Associate Prof.: Yoshimichi Andoh (CCS) →Implementation & parallelization

Dr. Tatsuya Sakashita → Implementation of solid harmonics, etc.

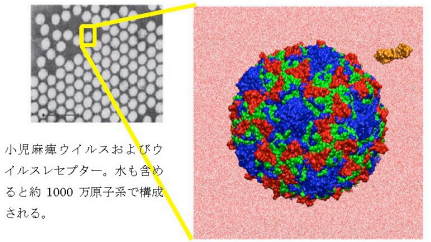
, and 8 post-docs; 3 Ph.D, 12 Master course, 5 undergraduate students

Research Subjects

- Plasma membranes of normal and cancerous cells
- Viruses, and their recognition by receptors
- Micelle formation and micellar solubilization
- Polymer material
- Development of theory of MD calculation, and MD simulation software, MODYLAS



粗視化シミュレーションによるマルチラメラベシクルの生成



マルチラメラベシクル、小児麻痺ウイルスの分子動力学シミュレーション

Molecular dynamics (MD) calculation

Basic equation :
Newton's equation of motion

$$m_i \frac{d^2 \boldsymbol{r}_i}{dt^2} = \boldsymbol{F}_i \qquad i = 1, \ldots, N$$

Numerical integration

Generalization
Verlet's method ⟶ RESPA[1]

$$\boldsymbol{v}_i\left(t + \frac{\Delta t}{2}\right) = \boldsymbol{v}_i(t) + \frac{\Delta t}{2}\frac{\boldsymbol{F}_i(\Delta t)}{m_i}$$

$$\boldsymbol{r}_i(t + \Delta t) = \boldsymbol{r}_i(t) + \Delta t\, \boldsymbol{v}_i\left(t + \frac{\Delta t}{2}\right)$$

$$\boldsymbol{v}_i\left(t + \Delta t\right) = \boldsymbol{v}_i\left(t + \frac{\Delta t}{2}\right) + \frac{\Delta t}{2}\frac{\boldsymbol{F}_i(t + \Delta t)}{m_i}$$

cycle

t : time
$\boldsymbol{r}_i$: atom coordinate
$\boldsymbol{F}_i$: forces working on each atom
m_i : atom mass
N : number of atoms O(10^4-10^7)

2nd order ordinary differential equations for $3N$ degree of freedoms
[initial conditions: $\boldsymbol{r}_i(0), \boldsymbol{v}_i(0)$]

$\boldsymbol{v}_i$: atom velocitiy

Δt : time step size O(10^{-15}) sec

100 ns long MD calculations = 10^8 cycles

[1] M. Tuckermann et al. J. Chem. Phys. 97, 1990 (1992).

Potential function

$$F_i = -\frac{\partial \Phi_{tot}(r_1,\ldots,r_N)}{\partial r_i}$$

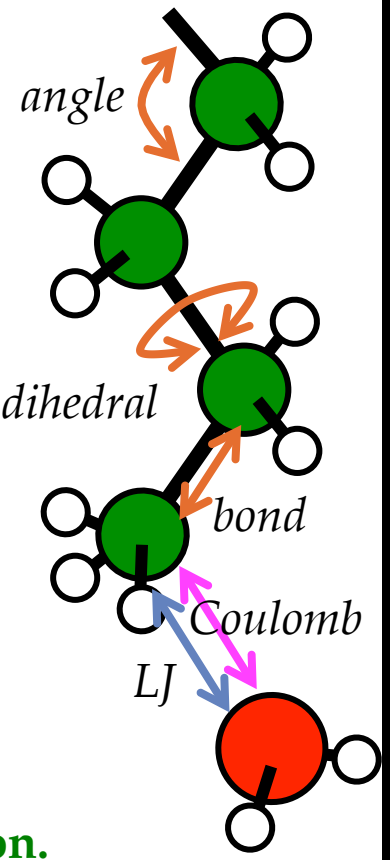
$\Phi_{tot}(r_1,\ldots,r_N)$: Total potential energy (Classical model)

Intra-molecular interactions

$$\Phi_{\mathrm{tot}} = \sum_{bonds} K_b(b-b_0)^2 + \sum_{angles} K_\theta(\theta-\theta_0)^2 + \sum_{dihedrals} K_\phi\left[1+\cos(n\phi-\delta)\right] + \sum_{impropers} K_\psi(\psi-\psi_0)^2 + \sum_{nonbonds}\left\{4\varepsilon_{ij}\left[\left(\frac{\sigma_{ij}}{r_{ij}}\right)^{12}-\left(\frac{\sigma_{ij}}{r_{ij}}\right)^{6}\right]+\frac{q_i q_j}{r_{ij}}\right\}$$

Lennard-Jones Interaction (Short range)

Coulombic interaction (Long range, $O(N^2)$)



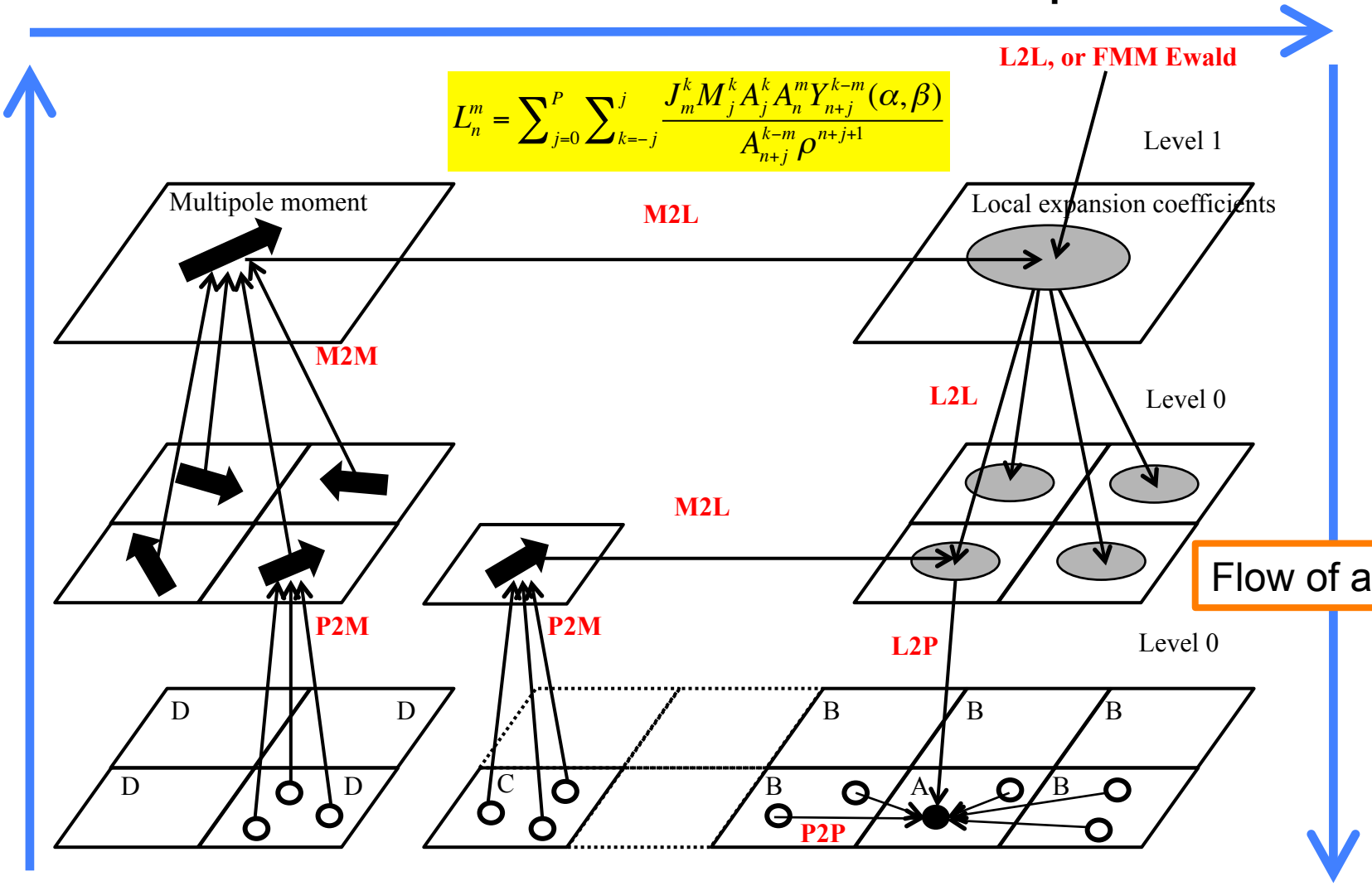
There is difficulty in effective MPI parallelization of this interaction.

We implemented the fast multipole method (FMM), besides particle mesh Ewald method, into MODYLAS

Review of FMM

- **O(*N*) algorithm**
- **suit for massive parallelization**

[2] L.F. Greengard (1987).



Flow of algorithm

P2M : $M_n^m = \sum_i q_i \rho_i Y_n^{-m}(\alpha_i, \beta_i)$

P2P: $\Phi_{\text{near}} = \sum_j \frac{q_j}{r_{ij}}$

L2P: $\Phi_{\text{far}} = \sum_{n=0}^{P} \sum_{m=-n}^{n} L_n^m Y_n^m(\theta,\phi) r^n$

M_n^m: multipole expansion coefficients, L_n^m: local expansion coefficients

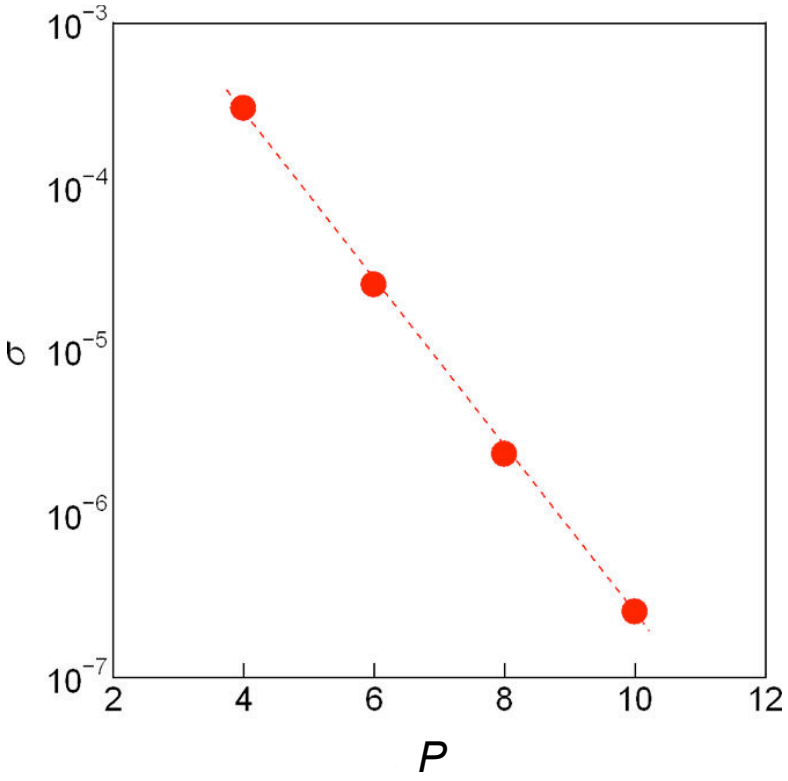
Accuracy of our coded FMM

The deviation of a force from the correct value obtained by the Ewald method:

$$\sigma_\alpha = \sqrt{\frac{\sum_i^N \left(F_{\alpha,i}^{P} - F_{\alpha,i}^{\mathrm{Ewald}}\right)^2}{\sum_i^N \left(F_{\alpha,i}^{\mathrm{Ewald}}\right)^2}} \qquad \alpha = x, y, z$$

P: The order of expansion, noted as "nmax" in the following slides

$$\Phi_{\mathrm{far}} = \sum_{n=0}^{P} \sum_{m=-n}^{n} L_n^m Y_n^m(\theta,\phi) r^n$$



Deviation σ (= σ_x = σ_y = σ_z) of the forces calculated by the FMM from the correct ones by the Ewald method.

[3] Y. Andoh, N. Yoshii, K. Fujimoto, et al., "MODYLAS: A highly parallelized general-purpose molecular dynamics simulation program for large-scale systems with long-range forces calculated by fast multipole method (FMM) and highly scalable fine-grained new parallel processing algorithms", J. Chem. Theory Comput., **2013**, 9, 3201.

Strong scaling test on the K computer

Measurement in March, 2012

N=10,000,000

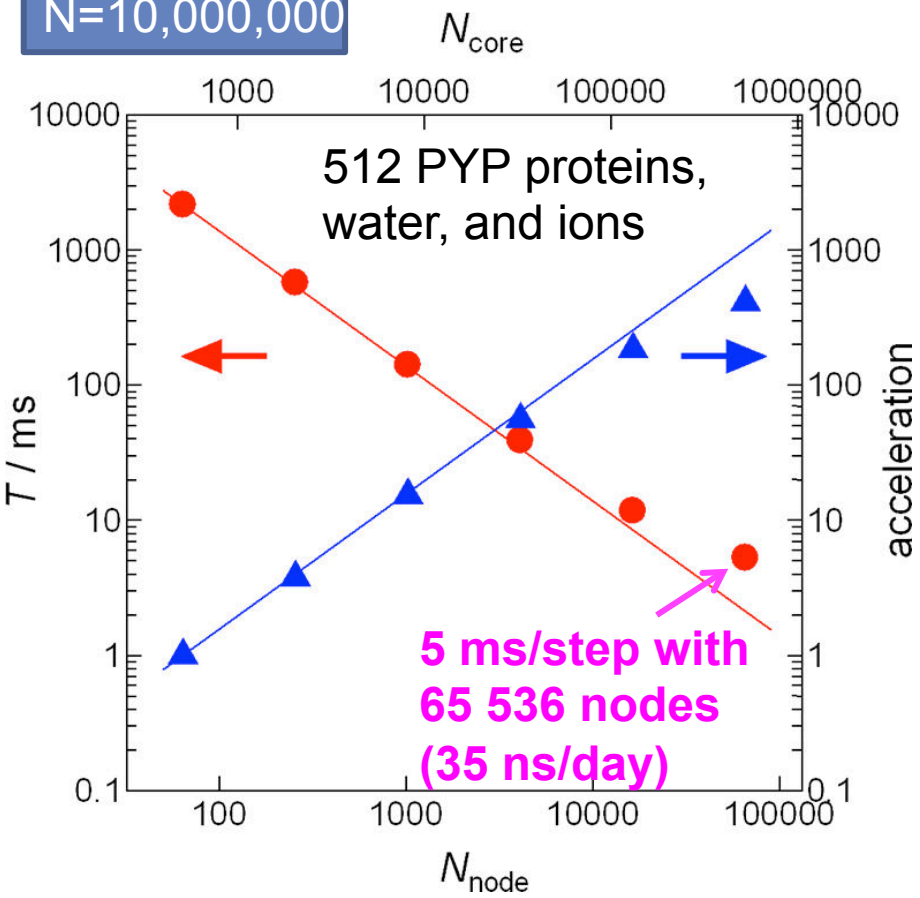


Fig. Measured overall calculation time per MD step (Δt) with respect to the 64-node calculation.

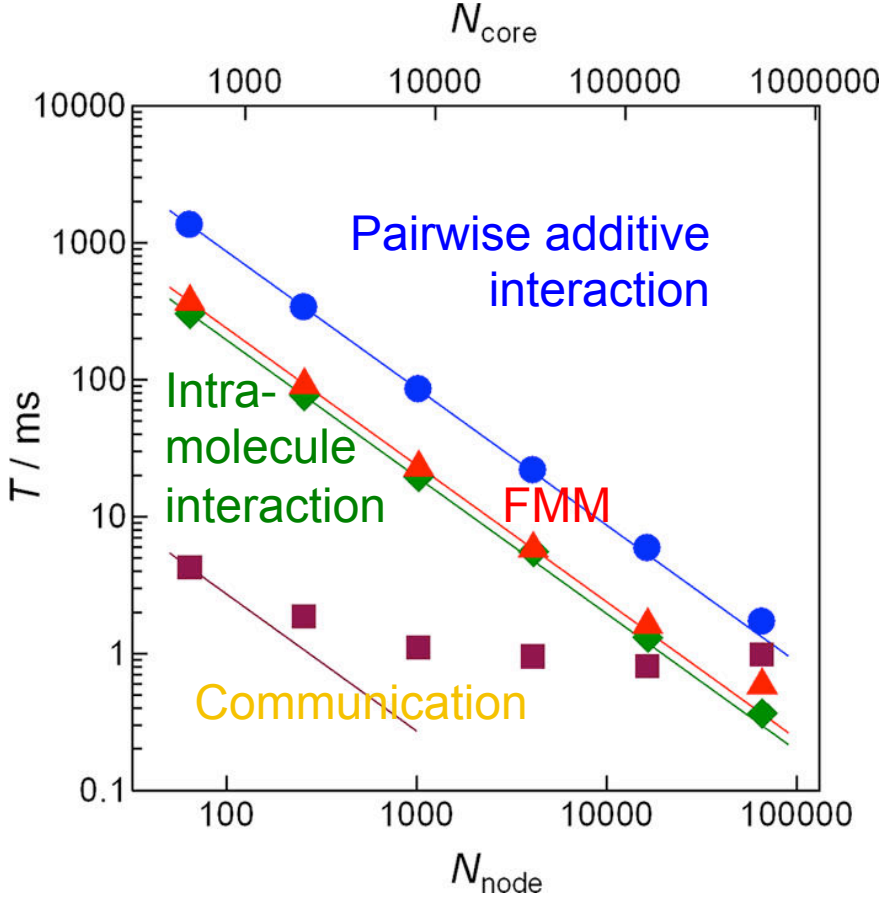
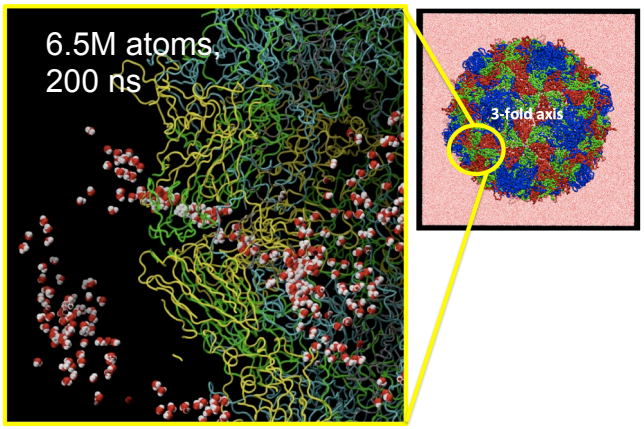


Fig. Measured partial calculation times and communication time per MD step (Δt)

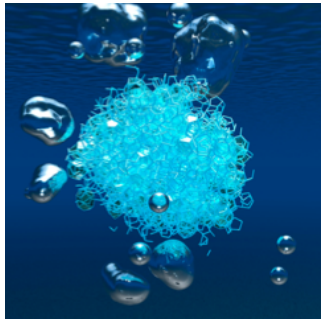
[3] Y. Andoh, N. Yoshii, K. Fujimoto, et al., "MODYLAS: A highly parallelized general-purpose molecular dynamics simulation program for large-scale systems with long-range forces calculated by fast multipole method (FMM) and highly scalable fine-grained new parallel processing algorithms", J. Chem. Theory Comput., **2013**, 9, 3201.

Applications of MODYLAS

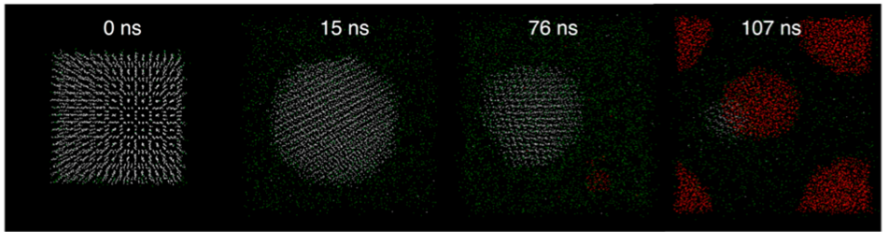


All-atomistic MD simulation of entire poliovirus empty capsid in solution
J. Chem. Phys. **141**, 165101 (2014).

Dissociation of methane hydrate (Okayama Univ.)
T.Yagasaki, M.Matsumoto, Y.Andoh, S.Okazaki, H.Tanaka,
J. Phys. Chem. B, **118**, 1900 (2014).
J. Phys. Chem. B,, **118**, 11797 (2014).;

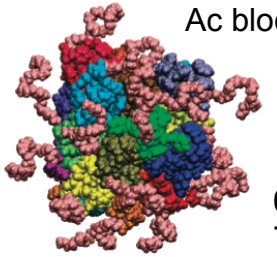


0.4 M atoms 120 ns



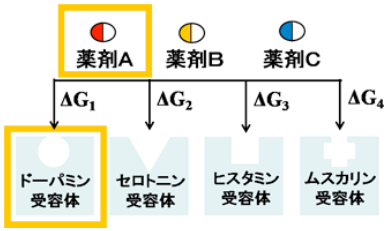
Examples of collaboration with companies

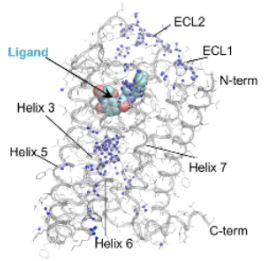
- Nippon-Kayaku
  J. Phys. Chem. B, **113**, 15181 (2009).
  Micelle of PEG-PBLG-Ac block copolymers



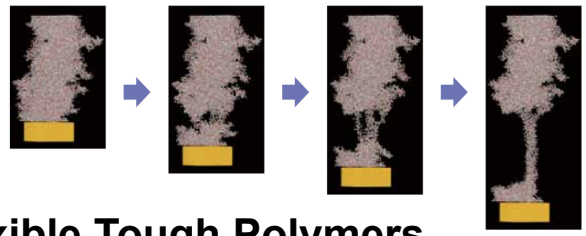
0.2 M atoms
7ns

- Dainihon Sumitomo Seiyaku
  PLoS ONE, **11**, 1 (2016).
  Drug-membrane protein affinity





- Nitto Denko
  Polymer adhesives



ImPACT Project (Prof. Ito):
Realizing Ultra-Thin and Flexible Tough Polymers

革新的研究開発推進プログラム ImPACT

MODYLAS is not benchmark software, but practical general-purpose MD software.

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- **Molecular dynamics calculation combined with FMM**
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Replacement of the spherical harmonics by solid harmonics
Reduction of arithmetic amount in FMM

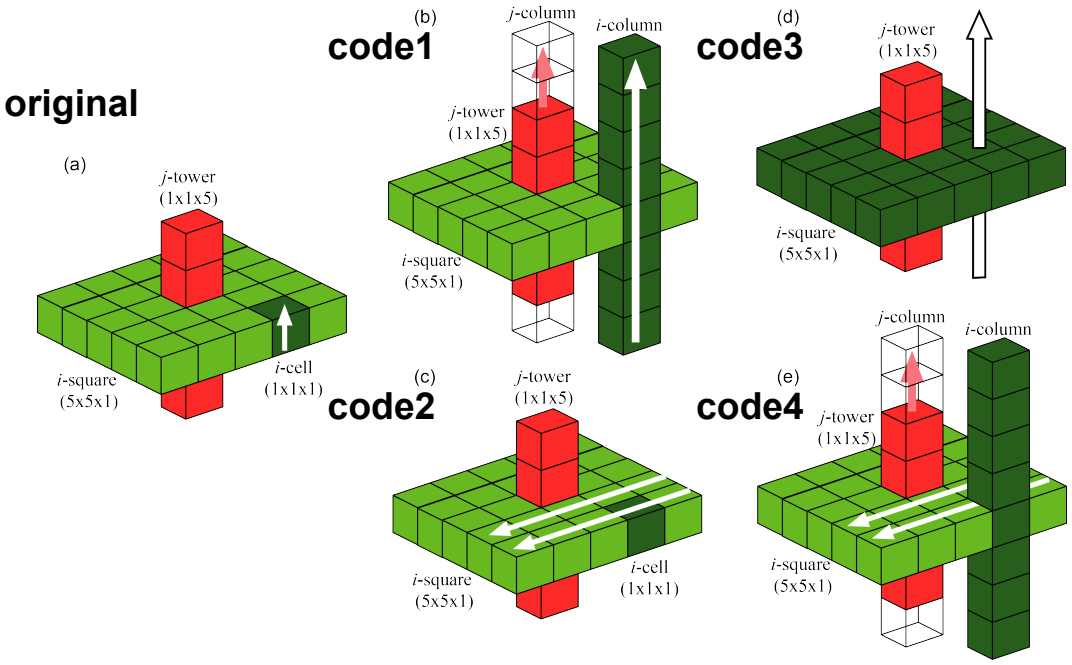
Functions:

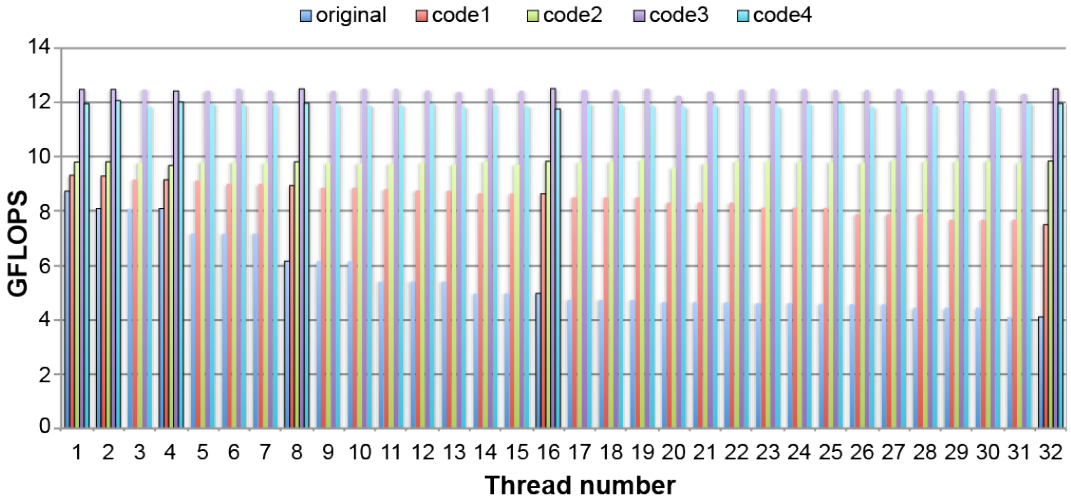
FMM for rectangular unit cell, with anisotropic cell division
Pressure tensor calculated by the FMM (J. Comput. Chem., in press)
Implementation of the regularized FMM
Implementation of bond-breaking potential for polymer simulation
Implementation of the EBV potential for proton-transfer simulation

- **Conclusion**

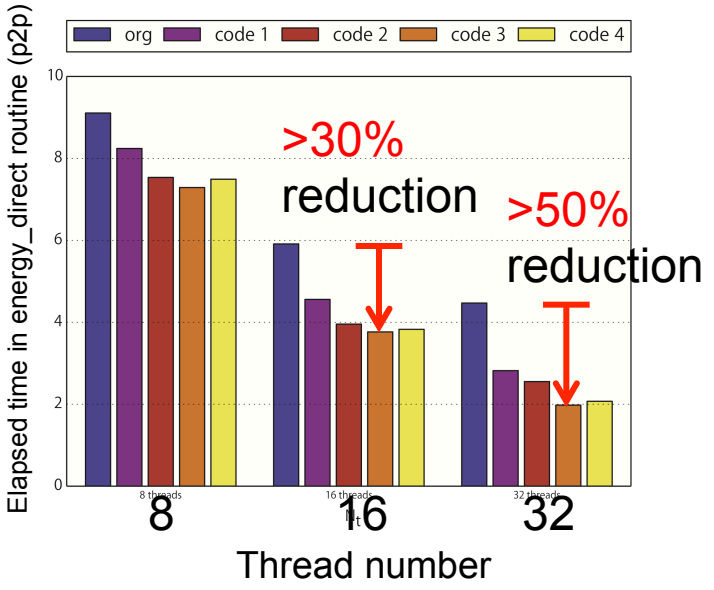
Red: today's topics

Performance:
1. Improvement thread-level parallelization (p2p)





New codes 1-4 realize,
- Load balancing between threads
- Enlargement of granularity for each thread



Y. Andoh, S. Suzuki, S. Ohshima, et al., J. of Supercomputing, in press

2. Replacement of the spherical harmonics by solid harmonics

P2M (Particle to Multipole, calculation of multipole moments)

$$M_\ell^m = \sum_{i=1}^{N} q_i \, R_\ell^m(\mathbf{r}_M - \mathbf{r}_i)$$

Regular solid harmonics

$$R_\ell^m(r,\theta,\phi) = \frac{r^\ell}{(\ell+m)!} P_\ell^m(\cos\theta)\, e^{im\phi}$$

M2M (Multipole to Multipole, to translate of expansion center of M)

$$M'^m_\ell = \sum_{\lambda=0}^{n_{\max}} \sum_{\mu=-\lambda}^{\lambda} M_{\ell-\lambda}^{m-\mu} R_\lambda^\mu(\mathbf{r}_{M'} - \mathbf{r}_M)$$

Traditional M2L with Y_n^m

$$L_n^m = \sum_{j=0}^{P} \sum_{k=-j}^{j} \frac{J_m^k M_j^k A_j^k A_n^m Y_{n+j}^{k-m}(\alpha,\beta)}{A_{n+j}^{k-m} \rho^{n+j+1}}$$

M2L (Multipole to Local expansion coeffs)

$$L_\ell^m = \sum_{\lambda=0}^{n_{\max}} \sum_{\mu=-\lambda}^{\lambda} M_\lambda^\mu \, S_{\ell+\lambda}^{-(m+\mu)}(\mathbf{r}_L - \mathbf{r}_M)$$

Singular solid harmonics

$$S_\ell^m(r,\theta,\phi) = (-1)^{\ell+m} \frac{(\ell-m)!}{r^{\ell+1}} P_\ell^m(\cos\theta)\, e^{im\phi}$$

L2L (Local expansion to Local expansion, to translate of expansion center of L)

$$L'^m_\ell = \sum_{\lambda=\ell}^{n_{\max}} \sum_{\mu=-\lambda}^{\lambda} L_\ell^m \, R_{\lambda-\ell}^{\mu-m}(\mathbf{r}_{L'} - \mathbf{r}_L)$$

$$\Phi_i(\mathbf{r}) = q_i \sum_{\ell=0}^{n_{\max}} \sum_{m=-\ell}^{\ell} L_\ell^m \, R_\ell^m(\mathbf{r} - \mathbf{r}_L)$$

$$\mathbf{F}_i = -q_i \sum_{\ell=0}^{n_{\max}} \sum_{m=-\ell}^{\ell} L_\ell^m \begin{bmatrix} \frac{1}{2}(R_{\ell-1}^{m+1} - R_{\ell-1}^{m-1}) \\ -\frac{1}{2}(R_{\ell-1}^{m+1} + R_{\ell-1}^{m-1}) \\ R_{\ell-1}^m \end{bmatrix}$$

Code has become simple greatly, make extension of FMM algorithm much easier

3. Reduction of arithmetic amount

Work by Dr. T. Sakashita & Prof. N. Yoshii

We have succeeded in reducing arithmetic amount of FMM by implementing the following three methods

(1) Utilizing symmetric nature of matrix element of multipole expansion coefficients *M* and local expansion coefficients *L*[4]

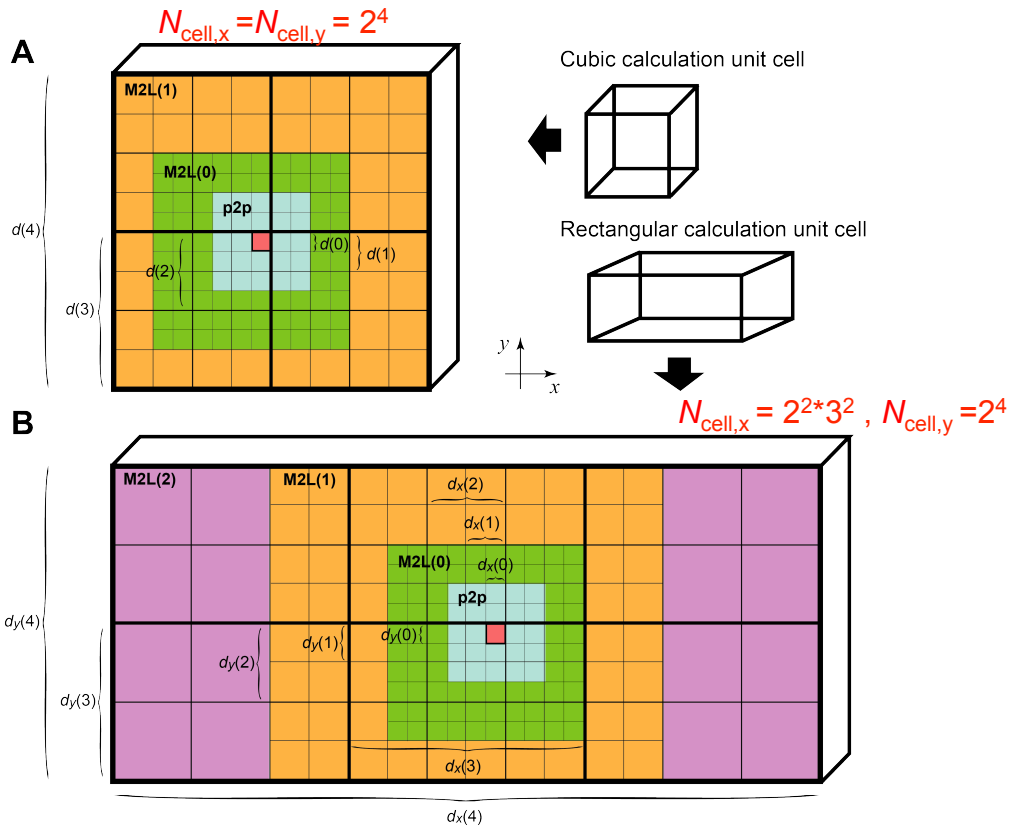
(2) Usage of recurrence formula[4] which can avoid a conversion from Cartesian coordinate to polar coordinate.

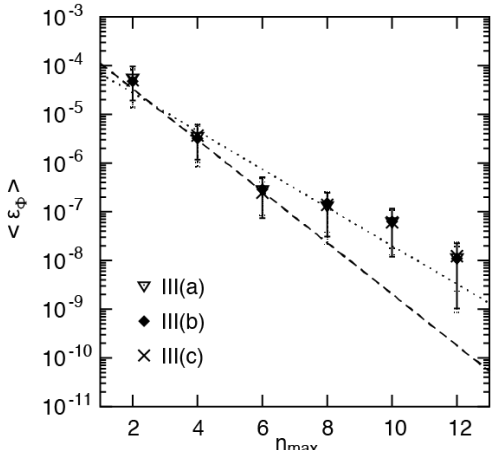
(3) Rotation of the axes in M2L operation[5]

[4] Basic idea is described in unpublished work by K. Nitadori, “Particle mesh multipole method: An efficient solver for gravitational/electrostatic forces based on multipole method and fast convolution over a uniform mesh”.

[5] C. A. White, M. Head-Gordon. J. Chem. Phys., Vol. 105, No. 12, pp. 5061– 5067, 1996.

Extension of function:
1. FMM for rectangular unit cell with anisotropic cell division





Error in potential vs nmax (reference value is obtained by Ewald method).
Error behavior strongly depends on style of cell division

Work by Y. Andoh & Prof. N. Yoshii

A: Traditional FMM:

- Cubic unit cell
- Isotropic cell division, following binary branches (octree structure)
- NPROCS = 2^n

B: Our extended FMM[6]:

- Rectangular unit cell
- Anisotropic cell division, following binary and ternary branches, independently selectable for each axis and each level
- NPROCS = 2^n*3^m is available

[6]Y. Andoh, N. Yoshii, S. Okazaki, *will be submitted soon.*

Extension of function:
2. New expression for pressure tensor in FMM

Work by Prof. N. Yoshii

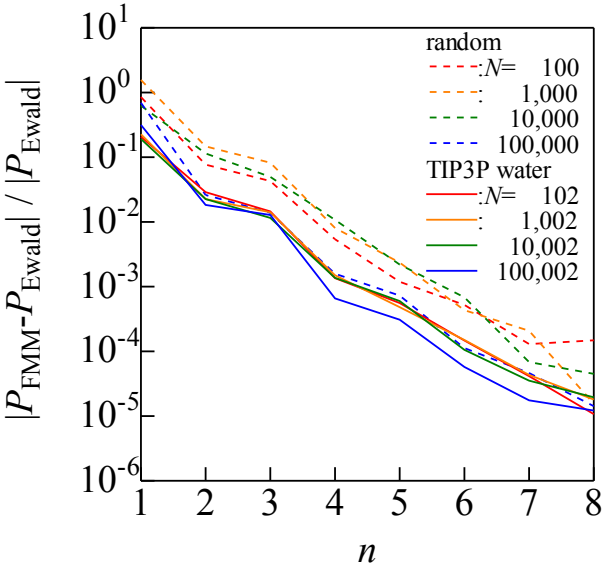
Pressure tensor

$$P_{\mathrm{FMM}\alpha\beta} = -\frac{1}{V}\sum_{\gamma}\frac{\partial \Phi_{\mathrm{FMM}}}{\partial h_{\alpha\gamma}} h_{\gamma\beta}$$

cf. Isotropic pressure (implemented in present MODYLAS 1.0.4)

$$P_{\mathrm{FMM}} = \frac{\Phi_{\mathrm{FMM}}}{3V}$$

transform	formula
P2M	$\frac{\partial M_n^m(l_{\max})}{\partial h_{\alpha\beta}} = \sum_{i=1}^{N} q_i \left[\nabla_i Y_n^{-m}(\hat{\boldsymbol{\rho}}_i)\rho_i^n\right]_\alpha s_{i\beta}$
M2M	$\frac{\partial M_j^k(l+1)}{\partial h_{\alpha\beta}} = \sum_{n=0}^{j}\sum_{m=-n}^{n} T_{\mathrm{MM},j,n}^{k,m}\left\{\frac{\partial M_{j-n}^{k-m}(l)}{\partial h_{\alpha\beta}} Y_n^{-m}(\hat{\boldsymbol{\rho}}_{\mathrm{MM}})\rho_{\mathrm{MM}}{}^n + M_{j-n}^{k-m}(l)\left[\nabla_{\mathrm{MM}} Y_n^{-m}(\hat{\boldsymbol{\rho}}_{\mathrm{MM}})\rho_{\mathrm{MM}}{}^n\right]_\alpha s_{\mathrm{MM}\beta}\right\}$
M2L	$\frac{\partial L_j^k(l)}{\partial h_{\alpha\beta}} = \sum_{n=0}^{\infty}\sum_{m=-n}^{n} T_{\mathrm{ML},j,n}^{k,m}\left\{\frac{\partial M_n^m(l)}{\partial h_{\alpha\beta}}\frac{Y_{n+j}^{m-k}(\hat{\boldsymbol{\rho}}_{\mathrm{ML}})}{\rho_{\mathrm{ML}}^{n+j+1}} + M_n^m(l)\left[\nabla_{\mathrm{ML}}\frac{Y_{n+j}^{m-k}(\hat{\boldsymbol{\rho}}_{\mathrm{ML}})}{\rho_{\mathrm{ML}}^{n+j+1}}\right]_\alpha s_{\mathrm{ML}\beta}\right\}$
M2L (image cells)	$\sum_{v\neq 0}^{v_{\max}}\frac{Y_n^m(\hat{\boldsymbol{\rho}}_v)}{\rho_v^{n+1}} = \sum_{v\neq 0}^{v_{\max}}\frac{Y_n^m(\hat{\boldsymbol{\rho}}_v)}{\rho_v^{n+1}}\frac{\Gamma(n+1/2,\kappa^2\rho_v^2)}{\Gamma(n+1/2)} + \sum_{H\neq 0}^{H_{\max}}\frac{i^m\pi^{n-1/2}Y_n^m(\hat{\boldsymbol{v}}_H)v_H^{n-2}\exp(-\pi^2 v_H^2/\kappa^2)}{V\Gamma(n+1/2)}$
M2L (image cells)	$\frac{\partial}{\partial h_{\alpha\beta}}\sum_{v\neq 0}^{v_{\max}}\frac{Y_n^m(\hat{\boldsymbol{\rho}}_v)}{\rho_v^{n+1}}\frac{\Gamma(n+1/2,\kappa^2\rho_v^2)}{\Gamma(n+1/2)} = \sum_{v\neq 0}^{v_{\max}}\frac{1}{\Gamma(n+1/2)}\left[\Gamma(n+1/2,\kappa^2\rho_v^2)\nabla_v\frac{Y_n^m(\hat{\boldsymbol{\rho}}_v)}{\rho_v^{n+1}} - 2\frac{\kappa^{n+1}e^{-\kappa^2\rho_v^2}Y_n^m(\hat{\boldsymbol{\rho}}_v)}{\rho_v^2}\boldsymbol{\rho}_v\right]_\alpha s_{v\beta}$
M2L (image cells)	$\frac{\partial}{\partial h_{\alpha\beta}}\sum_{H\neq 0}^{H_{\max}}\frac{i^m\pi^{n-1/2}Y_n^m(\hat{\boldsymbol{v}}_H)v_H^{n-2}\exp(-\pi^2 v_H^2/\kappa^2)}{V\Gamma(n+1/2)} = -\frac{i^m\pi^{n-1/2}v_H^{n-4}}{V\Gamma(n+1/2)}\exp\left(-\frac{\pi^2 v_H^2}{\kappa^2}\right)$ $\times\sum_{H\neq 0}^{H_{\max}}\left(Y_n^m(\hat{\boldsymbol{v}}_H)v_H^2 h_{\beta\alpha}^{-1} + \sum_\gamma v_{H\alpha}h_{\beta\gamma}^{-1}\left[Y_n^m(\hat{\boldsymbol{v}}_H)\left\{(n-2)-\frac{2\pi^2 v_H^2}{\kappa^2}\right\}v_{H\gamma} - \left(\boldsymbol{v}_H\times i\hat{\boldsymbol{L}}_H\right)_\gamma Y_n^m(\hat{\boldsymbol{v}}_H)\right]\right)$
M2L (image cells)	$\frac{\partial}{\partial h_{\alpha\beta}}\sum_{v\neq 0}^{v_{\max}}\frac{Y_{n+j}^{m-k}(\hat{\boldsymbol{\rho}}_v)}{\rho_v^{n+j+1}} = \sum_{v\neq 0}^{v_{\max}}\frac{Y_n^m(\hat{\boldsymbol{\rho}}_v)}{\rho_v^{n+1}}\frac{\Gamma(n+1/2,\kappa^2\rho_v^2)}{\Gamma(n+1/2)} + \frac{\partial}{\partial h_{\alpha\beta}}\sum_{H\neq 0}^{H_{\max}}\frac{i^m\pi^{n-1/2}Y_n^m(\hat{\boldsymbol{v}}_H)v_H^{n-2}\exp(-\pi^2 v_H^2/\kappa^2)}{V\Gamma(n+1/2)}$
M2L (image cells)	$\frac{\partial L_j^k(0)}{\partial h_{\alpha\beta}} = \sum_{n=0}^{\infty}\sum_{m=-n}^{n} T_{\mathrm{ML},j,n}^{k,m}\left\{\frac{\partial M_n^m(0)}{\partial h_{\alpha\beta}}\sum_{v\neq 0}^{v_{\max}}\frac{Y_{n+j}^{m-k}(\hat{\boldsymbol{\rho}}_v)}{\rho_v^{n+j+1}} + M_n^m(0)\left[\frac{\partial}{\partial h_{\alpha\beta}}\sum_{v\neq 0}^{v_{\max}}\frac{Y_{n+j}^{m-k}(\hat{\boldsymbol{\rho}}_v)}{\rho_v^{n+j+1}}\right]_\alpha s_{v\beta}\right\}$
L2L	$\frac{\partial L_j^k(l+1)}{\partial h_{\alpha\beta}} = \sum_{n=j}^{p}\sum_{m=-n}^{n} T_{\mathrm{LL},j,n}^{k,m}\left\{\frac{\partial L_n^m(l)}{\partial h_{\alpha\beta}} Y_{n-j}^{m-k}(\hat{\boldsymbol{\rho}}_{\mathrm{LL}})\rho_{\mathrm{LL}}{}^{n-j} + L_n^m(l)\left[\nabla_{\mathrm{LL}} Y_{n-j}^{m-k}(\hat{\boldsymbol{\rho}}_{\mathrm{LL}})\rho_{\mathrm{LL}}{}^{n-j}\right]_\alpha s_{\mathrm{LL}\beta}\right\}$
L2P	$P_{\alpha\beta} = -\frac{1}{V}\left\{\frac{1}{2}\sum_{j=0}^{p}\sum_{k=-j}^{j}\sum_\gamma\frac{\partial L_j^k(l_{\max})}{\partial h_{\alpha\gamma}}h_{\gamma\beta}\sum_{i=1}^{N}q_i Y_j^k(\hat{\boldsymbol{r}}_i)r_i^j - \frac{1}{2}\sum_{i=1}^{N}f_{i\alpha}r_{i\beta}\right\} + P_{\mathrm{pol}\alpha\beta}$



Parrinello-Rahman NPT ensemble MD
⇒anisotropic systems, e.g. crystal, membrane…

Physical property
⇒interfacial / surface tension …

[7] N. Yoshii, Y. Andoh, S. Okazaki, *J. Comput. Chem.*, in press.

Conclusion

- Our group have developed a general-purpose MD calculation software MODYLAS combined with FMM.

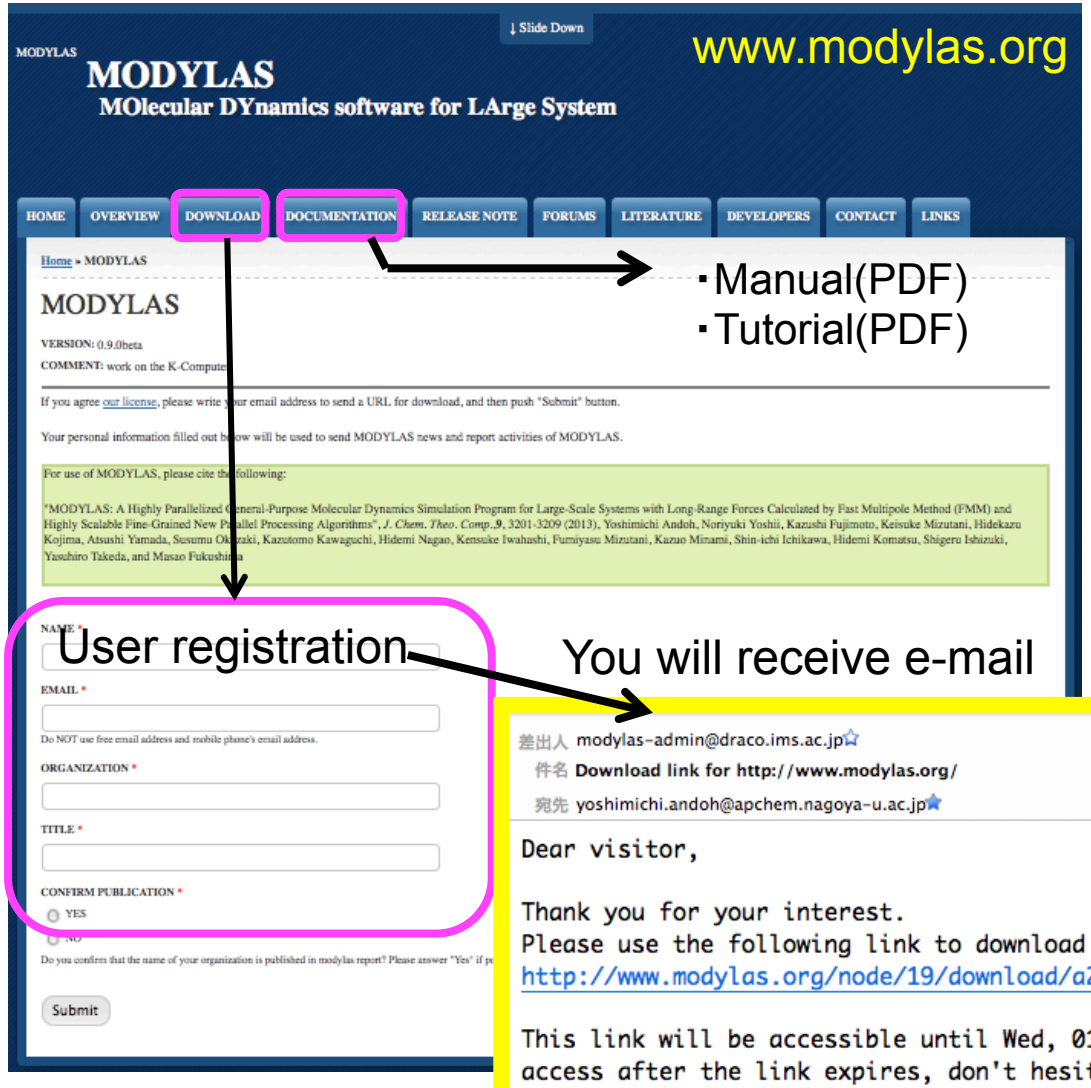
- MODYLAS has been creating remarkable output of scientific researches with the K computer, and its successor models (FX100).

- To realize large-scale MD calculation on the post K computer, we continue to make many efforts to improve both performance of arithmetic operation and MPI communication, and to extend functions of software.

- We need further efforts to achieve the goal

How to download MODYLAS

Full version of MODYLAS :



Miniapp version of MODYLAS :

http://fiber-miniapp.github.io/

FIBER

Code is open, but it is not “open source”.
Read LICENSE.pdf in package.

Related publications

Software developments:

[1] Y. Andoh, N. Yoshii, K. Fujimoto, et al., "MODYLAS: A highly parallelized general-purpose molecular dynamics simulation program for large-scale systems with long-range forces calculated by fast multipole method (FMM) and highly scalable fine-grained new parallel processing algorithms", *J. Chem. Theory Comput.*, **2013**, 9, 3201.
[2] Y. Andoh, S. Suzuki, S. Ohshima, T. Sakashita, M. Ogino, T. Katagiri, N. Yoshii, and S. Okazaki, "A thread-level parallelization of pairwise additive potential and force calculations suitable for current many-core architectures", *J. of Supercomputing,* in press
[3] N. Yoshii, Y. Andoh, and S. Okazaki, "Pressure tensor for electrostatic interaction calculated by fast multipole method with periodic boundary condition", *J. Comput. Chem.*, in press.
[4] Y. Andoh, N. Yoshii, and S. Okazaki, "Fast multipole method adaptive to a rectangular unit cell and partitioned tree structure with a mixture of binary and ternary branches", *will be submitted soon*.

Scientific outputs:

[5] Y. Andoh, N. Yoshii, A. Yamada, K. Fujimoto, H. Kojima, M. Mizutani, A. Nakagawa, A. Nomoto, S. Okazaki, "All-atom molecular dynamics simulation study of entire polivirus empty capsids in solution", J. Chem. Phys., 141, 165101 (2014).
[6] T.Yagasaki, M.Matsumoto, Y.Andoh, S.Okazaki, H.Tanaka, "Effect of bubble formation on the dissociation of methane hydrate in water: A molecular dynamics study", *J. Phys. Chem. B*, **118**, 1900 (2014).
[7] T.Yagasaki, M.Matsumoto, Y.Andoh, S.Okazaki, H.Tanaka, "Dissociation of methane hydrate in aqueous NaCl solutions", *J. Phys. Chem. B*,, **118**, 11797 (2014).;