

Determination of Proper Time Step for Molecular Dynamics Simulation

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Received February 28, 2000

In this study we have investigated the determination of proper time step in molecular dynamics simulation. Since the molecular dynamics is mathematically related to nonlinear dynamics, the analysis of eigenvalues is used to explain the relationship between the time step and dynamics. The tracings of H₂ and CO₂ molecular dynamics simulation agrees very well with the analytical solutions. For H₂, the time step less than 1.823 fs provides stable dynamics. For CO₂, 3.808 fs might be the maximum time step for proper molecular dynamics. Although this results were derived for most simple cases of hydrogen and carbon dioxide, we could quantitatively explain why improperly large time step destroyed the molecular dynamics. From this study we could set the guide line of the proper time step for stable dynamics simulation in molecular modeling software.

Introduction

Molecular dynamics^{1,2} is a simulation³ method describing the molecular motion by the equation of motion which Newton's classical mechanics describes. The first computer simulation using molecular dynamics method was performed on simple fluids, later on the solution of simple molecules like water.^{4,5} Later, according to the acceleration of development of computation speed and storage capacity, molecular dynamics of bigger molecules and biopolymers have been performed.⁶

In the molecular dynamics simulation, the atoms included in molecules move according to the Newtonian equation of motions. In other words, when we know the structure of molecule at one moment, we can find the structure at the next moment using the integration method of the equation of motion with time starting from here. The process of integrating the equation of motion can be achieved by several kinds of algorithm. The commonly important concept in the various algorithms is the time step (Δt). If a big time step is used, the motion of molecule becomes unstable due to the very big error occurring in the integration. Therefore, molecules may not have a normal structure any more. Reversely, if a very small value of time step is used, it will not be efficient due to a very long calculation time.⁷

Therefore, the selection of the time step is very important to perform the molecular dynamics simulation. In normal molecular dynamics simulation, we use 1 femto second (fs: 10⁻¹⁵ sec) time step. There are some logical explanations^{8,9} about the usage of 1 fs time step. One of the explanations is that stable dynamics will be executed only if we use the smaller time step compared to the period of the highest vibrational frequency of the molecule. If we can determine the biggest time step for a stable dynamics, it is expected that the efficiency of the molecular dynamics simulation will be maximized.

In this paper, we have investigated the reasons why the dynamics sometimes break down after a maximum limit of time step in the simulation, and determined the exact point where a chaotic behavior start in molecular dynamics simulations of simple system such as hydrogen or carbon dioxide molecule.

Theory

If the energy of a molecule is expressed in a function of the molecular structure, the force acting on an atom can be determined by differentiating the energy with position of the atom A. (See Figure 1 for notations in Eq. (1)).

$$a_{xA} = \frac{dv_{xA}}{dt} = \frac{F_{xA}}{m_A} = -\frac{1}{m_A} \frac{\partial E}{\partial x_A} \quad (1)$$

where a_{xA} is the acceleration of atom A in x direction, v_{xA} the velocity of atom A in x direction, F_{xA} the force acting on atom A in x direction, E the potential energy of the molecule.

If the acceleration is calculated by the above method, we can determine the velocity by the integration of acceleration with time, and also the change of position by the integration of velocity with time. Several algorithms are proposed to carry out the integration processes. The most frequently used algorithm is called Verlet.³

$$v_{\frac{1}{2}} = v_{\frac{1}{2}} + a_0 \Delta t \quad (2)$$

$$x_1 = x_0 + v_{\frac{1}{2}} \Delta t \quad (3)$$

$$v_{\frac{3}{2}} = v_{\frac{1}{2}} + a_1 \Delta t \quad (4)$$

$$x_2 = x_1 + v_{\frac{3}{2}} \Delta t \quad (5)$$

.....

The other popular one is leap-frog³ algorithm. This gives virtually the same result as Verlet algorithm as following.

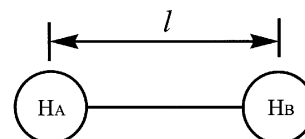


Figure 1. The Hydrogen Molecule. H_A : hydrogen atom A, H_B : hydrogen B, l : bond length between two atoms.

$$v_1 = 2v_0 + a_0(\Delta t) \quad (6)$$

$$x_1 = x_0 + v_1(\Delta t) \quad (7)$$

$$x_1 = 2x_0 - x_{-1} + a_0(\Delta t)^2 \quad (8)$$

...

$$x_2 = 2x_1 - x_0 + a_1(\Delta t)^2 \quad (9)$$

.....

In this study, we investigated the changes of molecular dynamics by the different time-step (Δt). The simpler trial molecules are hydrogen (H_2) and carbon dioxide (CO_2).

Mathematical Model. Let V be a partial group of R^2 . If a certain function F is described as $F: V \rightarrow R^2$, this function is called "map". This kind of function F can be expressed by the following form.

$$F(V) = \begin{pmatrix} f(v) \\ g(v) \end{pmatrix} \text{ for all } v \text{ in } V \quad (10)$$

Here, f and g are the coordinate functions of F . If this coordinate function is internal coordinate system of a molecule, the process of molecular dynamics simulation is same as the iterative calculations of the above defined function like following procedure.

$$v_1 = F(v_0) = \begin{pmatrix} f(v_0) \\ g(v_0) \end{pmatrix} \quad (11)$$

$$v_2 = F(v_1) = \begin{pmatrix} f(v_1) \\ g(v_1) \end{pmatrix} \quad (12)$$

...

The movements of $v_0, v_1, v_2, \dots, v_n$ from the above iterative calculations describe the same results as molecular dynamics simulation. In order to analyse the dynamics of molecular system as molecular dynamics simulation, one should know the exact variations of $v_0, v_1, v_2, \dots, v_n$ and would define the partial derivatives of the above defined functions.

$$DF(v) = \begin{pmatrix} \frac{\partial f}{\partial x}(v) & \frac{\partial f}{\partial y}(v) \\ \frac{\partial g}{\partial x}(v) & \frac{\partial g}{\partial y}(v) \end{pmatrix} \quad (13)$$

This kind of partial derivative can be expressed in terms of matrix. The dynamics depend upon the outcome (real or complex number and the sign of imaginary part for complex number) of the eigenvalues of the matrix. Therefore, we have defined $DF(v)$ for simple molecular systems (H_2 and CO_2) and analyzed the outcome of the eigenvalues for different time-step (Δt) for molecular dynamics simulations.

Hydrogen Molecule (Harmonic oscillator function). The potential energy of the hydrogen molecule can be expressed to the following equation.

$$E = \frac{k_s}{2}(l-l^0)^2 \quad (14)$$

where l is the bond length, l^0 the most stable bond length, k_s the force constant.

In the above equation, l^0 is 0.64 Å in MM2 force field.¹⁰ The force constant is 5 millidyne/Å (= 500 N/m). The mass of the hydrogen atom is 1.66113×10^{-27} kg. The analytical method of the molecular dynamics simulation for the hydrogen molecule using the above equation is following.

$$E = \frac{k_s}{2}(l-l^0)^2 \quad (15)$$

where $l = x_B - x_A$ ($x_B > x_A$), x_A and x_B are the positions of H_A and H_B .

The positions of the hydrogen atoms by the leap-frog algorithm will be changed by the following equations.

$$x_{A2} = 2x_{A1} - x_{A0} + a_{A1}(\Delta t)^2 \quad (16)$$

$$x_{B2} = 2x_{B1} - x_{B0} + a_{B1}(\Delta t)^2 \quad (17)$$

Therefore, the bond length defined by $l = x_B - x_A$, will be

$$l_2 = 2l_1 - l_0 - (a_{A1} - a_{B1})(\Delta t)^2 \quad (18)$$

The accelerations of atoms A and B are

$$a_A = -\frac{1}{m_A} \frac{\partial E}{\partial x_A} = -\frac{1}{m_A} \frac{\partial}{\partial x_A} \frac{k_s}{2}(x_B - x_A - l^0)^2 = \frac{k_s}{m_A}(l - l^0) \quad (19)$$

$$a_B = -\frac{k_s}{m_B}(l - l^0) \quad (20)$$

Then, the Eq. (18) will be converted to Eq. (21).

$$\begin{aligned} l_2 &= 2l_0 - l_0 - \left(\frac{1}{m_A} - \frac{1}{m_B} \right) k_s (l_1 - l^0) (\Delta t)^2 \\ &= 2l_1 - l_0 - \frac{k_s}{\mu} (l_1 - l^0) (\Delta t)^2 \end{aligned} \quad (21)$$

where μ is the reduced mass (= 8.3057×10^{-28} kg)

Here, we define a vector containing the current bond length (l_n) and the previous bond length (l_{n-1}) before the time step Δt .

$$\begin{pmatrix} l_{n-1} \\ l_n \end{pmatrix} \quad (22)$$

Then, the above Eq. (21) can be transformed to the following form,

$$\begin{pmatrix} l_1 \\ l_2 \end{pmatrix} = \begin{pmatrix} l_1 \\ 2l_1 - l_0 - \frac{k_s}{\mu} (l_1 - l^0) (\Delta t)^2 \end{pmatrix} \quad (23)$$

If we rewrite the function with a general expression,

$$F \begin{pmatrix} x \\ y \end{pmatrix} = \begin{pmatrix} y \\ 2y - x - \frac{k_s}{\mu}(y - l^0)(\Delta t)^2 \end{pmatrix} \quad (24)$$

Therefore, the simulation of molecular dynamics for the hydrogen molecule is the same as the repeated iterations of the Eq. (24). In order to understand the dynamics, we should know the orbit and should define the differential of the function.

$$DF \begin{pmatrix} x \\ y \end{pmatrix} = \begin{pmatrix} 0 & 1 \\ -1 & 2 - \frac{k_s}{\mu}(\Delta t)^2 \end{pmatrix} \quad (25)$$

Since the molecular dynamics is mathematically related to nonlinear dynamics,¹¹ the analysis of eigenvalues is used to explain the relationship between the time step and dynamics.¹² If find the eigenvalue of the above matrix, the dynamics of the Eq. (24) will be completely understood. If we define the eigenvalue λ , λ should satisfy the following determinant.

$$\begin{vmatrix} -\lambda & 1 \\ -1 & 2 - \frac{k_s}{\mu}(\Delta t)^2 - \lambda \end{vmatrix} \quad (26)$$

Therefore,

$$\lambda^2 + \left(\frac{k_s}{\mu}(\Delta t)^2 - 2 \right) \lambda + 1 = 0 \quad (27)$$

Depending on the size of Δt , the roots (λ) satisfying the Eq. (27) can be several kinds of complex numbers.

i) λ 's are complex roots and the real parts of the roots are positive.

The following condition should be satisfied for λ to be complex numbers.

$$\left(\frac{k_s}{\mu}(\Delta t)^2 - 2 \right)^2 - 4 < 0 \quad (28)$$

Therefore, the range of Δt for to be complex roots is

$$0 < \Delta t < 2 \left(\frac{\mu}{k} \right)^{1/2} \quad (29)$$

And the condition that the real parts of the roots are positive will require the following range of Δt .

$$-\left(\frac{k_s}{\mu}(\Delta t)^2 - 2 \right) > 0 \quad (30)$$

$$-\left(\frac{2\mu}{k} \right)^{1/2} < \Delta t < \left(\frac{2\mu}{k} \right)^{1/2} \quad (31)$$

The time-step Δt simultaneously satisfying the above two conditions should be:

$$0 < \Delta t < \left(\frac{2\mu}{k} \right)^{1/2} \approx 1.823 \times 10^{-15} \quad (32)$$

ii) λ 's are complex roots and the real parts of the roots are

Table 1. The eigenvalues and turning points for H₂ dynamics

Cases	λ 's	Maximum Δt (sec)
1	2 complex numbers, real part (+)	1.823×10^{-15}
2	2 complex numbers, real part (-)	2.578×10^{-15}
3	2 real numbers	∞

negative.

The similar process gives the following range of time-step:

$$1.823 \times 10^{-15} \approx \left(\frac{2\mu}{k} \right)^{1/2} < \Delta t < \left(\frac{2\mu}{k} \right)^{1/2} \approx 2.578 \times 10^{-15} \quad (33)$$

ii) λ 's are real roots

$$\Delta t > 2 \left(\frac{\mu}{k} \right)^{1/2} \approx 2.578 \times 10^{-15} \quad (34)$$

Carbon Dioxide (CO₂). The potential energy of carbon dioxide molecule is expressed like following.

$$E = \frac{k_s}{2}(l_a - l^0)^2 + \frac{k_s}{2}(l_b - l^0)^2 + \frac{k_\theta}{2}(\theta - \theta^0)^2 \quad (35)$$

$$\text{where : } l_a = x_C - x_{OA} \\ l_b = x_{OB} - x_C$$

The third term in above equation is related with an angle bending. The magnitude of contribution is determined to be very small to the total energy, and will be omitted in next calculation step for a simplification.

$$E \approx \frac{k_s}{2}(l_a - l^0)^2 + \frac{k_s}{2}(l_b - l^0)^2 \\ = \frac{k_s}{2}(x_C - x_{OA} - l^0)^2 + \frac{k_s}{2}(x_{OB} - x_C - l^0)^2 \quad (36)$$

For carbon dioxide, the force constant (k_s) of bond-stretching is 10 millidyne/Å, the equilibrium bond length (λ_0) is 1.162. The mass of carbon atom 1.99336×10^{-26} kg, and the mass of oxygen atom is 2.65781×10^{-26} kg.

Using the energy expression of carbon dioxide, we can calculate the forces acting on the individual atoms by differentiation of the energy with respect to the coordinate of each atom. From these forces, we can determine the accelerations of atoms.

$$a_{OA} = -\frac{1}{m_O} \frac{\partial E}{\partial x_{OA}} = \frac{k_s}{m_O}(l_a - l^0) \quad (37)$$

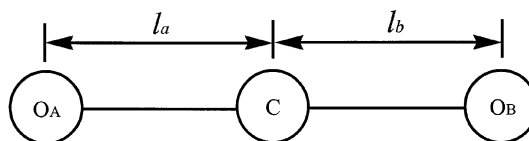


Figure 2. Carbon Dioxide Molecule. C: carbon atom, O_A and O_B: oxygen atoms, l_a : bond length between carbon and oxygen A, l_b : bond length between carbon and oxygen B.

$$a_C = -\frac{1}{m_C} \frac{\partial E}{\partial x_C} = -\frac{k_s}{m_C} ((l_a - l^0) - (l_b - l^0)) \quad (38)$$

$$a_{OB} = -\frac{1}{m_O} \frac{\partial E}{\partial x_{OB}} = -\frac{1}{m_O} (l_b - l^0) \quad (39)$$

Applying the acceleration of each atom to the Verlet algorithm, the following expressions of atomic coordinates after a time-step (Δt) can be obtained.

$$x_{OA}(t + \Delta t) = 2x_{OA}(t) - x_{OA}(t - \Delta t) + \frac{k_s}{m_O} (l_a - l^0) \Delta t^2 \quad (40)$$

$$x_C(t + \Delta t) = 2x_C(t) - x_C(t - \Delta t) - \frac{k_s}{m_C} ((l_a - l^0) - (l_b - l^0)) \Delta t^2 \quad (41)$$

$$x_{OB}(t + \Delta t) = 2x_{OB}(t) - x_{OB}(t - \Delta t) + \frac{k_s}{m_O} (l_b - l^0) \Delta t^2 \quad (42)$$

If one define $l_a = x_C - x_{OA}$, $l_b = x_{OB} - x_C$, the following equations can be obtained.

$$l_a(t + \Delta t) = 2l_a(t) - l_a(t - \Delta t) - \frac{k_s}{\mu} (l_a(t) - l^0) \Delta t^2 + \frac{k_s}{m_C} (l_b(t) - l^0) \Delta t^2 \quad (43)$$

$$l_b(t + \Delta t) = 2l_b(t) - l_b(t - \Delta t) - \frac{k_s}{\mu} (l_b(t) - l^0) \Delta t^2 + \frac{k_s}{m_C} (l_a(t) - l^0) \Delta t^2 \quad (44)$$

In order to use the similar vector expression as hydrogen molecule, one can define $w = l_a(t - \Delta t)$, $x = l_a(t)$, $y = l_b(t - \Delta t)$, $z = l_b(t)$. Then the above equations can be rewritten as following.

$$F \begin{pmatrix} w \\ x \\ y \\ z \end{pmatrix} = \begin{pmatrix} x \\ 2x - w - \frac{k_s}{\mu} (x - l^0) \Delta t^2 + \frac{k_s}{m_C} (z - l^0) \Delta t^2 \\ z \\ 2z - y - \frac{k_s}{\mu} (z - l^0) \Delta t^2 + \frac{k_s}{m_C} (x - l^0) \Delta t^2 \end{pmatrix} \quad (45)$$

In order to analyze the above function, one should derive the following partial derivative matrix.

$$DF \begin{pmatrix} w \\ x \\ y \\ z \end{pmatrix} = \begin{pmatrix} 0 & 1 & 0 & 0 \\ -1 & 2 - \frac{k_s}{\mu} \Delta t^2 & 0 & \frac{k_s}{m} \Delta t^2 \\ 0 & 0 & 0 & 1 \\ 0 & \frac{k_s}{\mu} \Delta t^2 & -1 & 2 - \frac{k_s}{\mu} \Delta t^2 \end{pmatrix} \quad (46)$$

The above matrix has four eigenvalues (λ) and the outcome will depend on the different time step (Δt).

ii) All four eigenvalues are complex numbers and the signs of real parts are plus.

$$0 < \Delta t < \left(\frac{2}{k \left(\frac{1}{m_O} + \frac{2}{m_C} \right)} \right)^{1/2} \approx 3.80751 \times 10^{-15} \quad (47)$$

ii) All four eigenvalues are complex numbers and the signs of real parts are minus for two of them.

$$\left(\frac{2}{k \left(\frac{1}{m_O} + \frac{2}{m_C} \right)} \right)^{1/2} < \Delta t < 2 \left(\frac{1}{k \left(\frac{1}{m_O} + \frac{2}{m_C} \right)} \right)^{1/2} \approx 5.38464 \times 10^{-15} \quad (48)$$

iii) Two eigenvalues are complex numbers and the signs of real parts are plus, and the remain two eigenvalues are real numbers.

$$2 \left(\frac{1}{k_s \left(\frac{1}{m_O} + \frac{2}{m_C} \right)} \right)^{1/2} < \Delta t < \left(\frac{2m_O}{k_s} \right)^{1/2} \approx 7.29083 \times 10^{-15} \quad (49)$$

iv) Two eigenvalues are complex numbers and the signs of real parts are minus, and the remain two eigenvalues are real numbers.

$$\left(\frac{2m_O}{k_s} \right)^{1/2} < \Delta t < 2 \left(\frac{m_O}{k_s} \right)^{1/2} \approx 10.3108 \times 10^{-15} \quad (50)$$

v) All four eigenvalues are real numbers

$$\Delta t > 2 \left(\frac{m_O}{k_s} \right)^{1/2} \approx 10.3108 \times 10^{-15} \quad (51)$$

Results and Discussion

In order to determine the proper time step (Δt) for molecular dynamics simulation, we have tried to find the turning points of dynamics through analytical calculation of leap-frog algorithm.

We have performed two simple cases for molecular dynamics computer simulation to prove that the calculated turning points are the drastically changing dynamics such as diverging bond distance.

Hydrogen Molecule. The potential energy function of hydrogen molecule is derived from MM2 force field¹⁰ and the molecular dynamics simulation is performed by leap-frog

Table 2. The eigenvalues and turning points for CO₂ dynamics

Cases	λ 's	Maximum Δt (sec)
1	4 complex numbers, real parts (+)	3.808×10^{-15}
2	4 complex numbers, 2 real parts (+)	5.385×10^{-15}
3	2 complex numbers, real parts (+)	7.291×10^{-15}
4	2 complex numbers, real parts (-)	10.311×10^{-15}
5	4 real numbers	∞

algorithm. The source code for the simulation is written by Visual Basic.

Since the purpose of this simulation is the recording of the bond-distance dynamics according to the time step (Δt), we plotted the 500 iterations of molecular dynamics for each time step from 0.00 to 3.00 femto seconds for 0.01 fs intervals. The tracings are accumulated in Figure 3.

In Figure 3, the horizontal axis is the time step and the vertical axis is the distance between two hydrogen atoms. The two vertical lines at 1.823 and 2.578 fs are at the values calculated from the analytical method (Table 1). For H₂, the time step less than 1.823 fs would provide stable dynamics. Onto this time step, the bond distance normally oscillates between +0.06 and -0.06 Angstrom from the average distance (0.64 Å) for molecular dynamics simulation. After 1.823 fs time step, the bond distance (H-H) begins to oscillate with much bigger amplitude until 2.578 fs where the molecular dynamics completely breaks down to go for a chaotic behavior.¹¹ As one compares the eigenvalues of Table 1 with the boundary values of Figure 3, the 2.578 fs calculated from theory is exactly matched for the time step of MD simulation. After this point (2.578 fs), the bond distance will jump from a certain value to another without continuity or physical meaning.

The stability of the numerical integration with respect to the time step can be tested directly by integrating the forces used by dynamics and comparing the integral with the analytical energy.⁹

In Figure 4, Integrated energy calculated numerically from a dynamics trajectory of hydrogen atoms in hydrogen molecule is compared with the analytical energy curve (harmonic form: $E = k_s(l - l^0)^2/2$). The time step used is 1 fs at a temperature of 300 K. The temperature is set by assigning an initial velocity of 1500 m/sec (0.015 Ånm/fs) to one of hydrogens along the vector connecting them. As two atoms approach each other, the numerical energy is higher than the analytical energy when the bond length is longer than equilibrium distance. The numerical value is lower when the

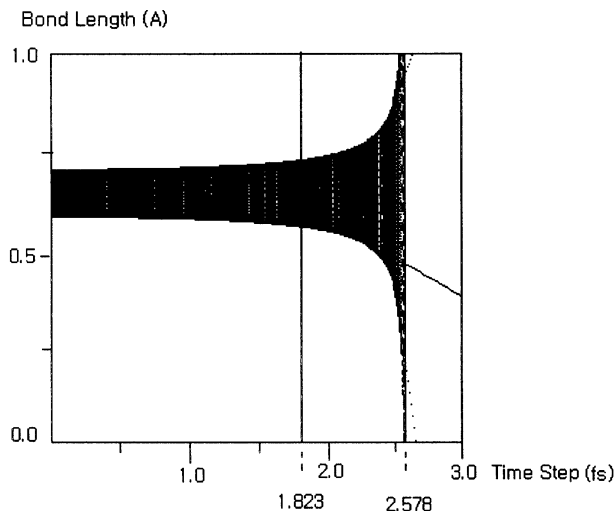


Figure 3. MD orbit diagram of H₂ molecule.

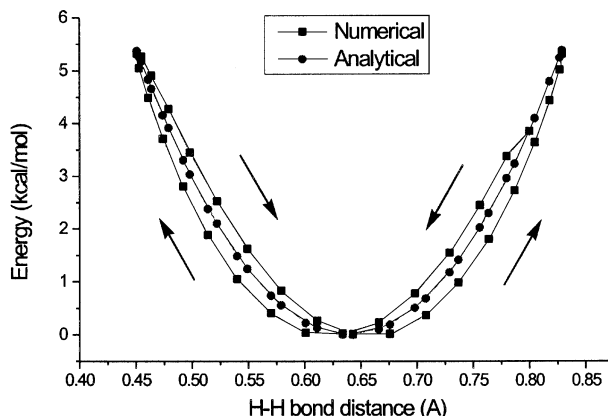


Figure 4. Numerical Integration of Energy from Molecular Dynamics of H₂ Molecule, 1 fs Time Step. Integrated energy calculated numerically from a dynamics trajectory of hydrogen atoms (squares) is compared with the analytical energy curve (circles).

bond length is shorter than equilibrium distance. However, after atoms collide, the integration energy is higher than the analytical energy when the bond length is shorter than equilibrium distance. Due to the assumption of harmonic form of energy, the numerical energy oscillates between maximum and minimum bond distance. The situation is different from the result of reference 9, where the hydrogen atoms have no bonding between them and the expression for analytical energy is Morse form: $E = D_b(1 - e^{-a(b-b_0)^2})$.

Carbon Dioxide (CO₂). A similar molecular dynamics simulation of carbon dioxide is also performed.

In Figure 5, four vertical lines at 3.808, 5.385, 7.291, and 10.311 fs are determined from the analytical solution. For CO₂, 3.808 fs might be the maximum time step for proper molecular dynamics. Onto this time step, the bond distance normally oscillates between +0.12 and -0.12 Ånm from the average distance (1.162 Å) for molecular dynamics simulation. After 3.808 fs time step, the bond distance begins to oscillate with much bigger amplitude until 5.385 fs where the molecular dynamics completely breaks down to go for a chaotic behavior. As one compares the eigenvalues of Table 2 with the boundary values of Figure 3, the 5.385 fs calculated from theory is exactly matched for the time step of MD simulation. After this point (5.385 fs), the bond distance will jump from a certain value to another without continuity.

The tracings of H₂ and CO₂ molecular dynamics simulation agree very well with the analytical solutions. For H₂, the time step less than 1.823 fs is providing stable dynamics. For CO₂, 3.808 fs might be the maximum time step for proper molecular dynamics.

Conclusion

In this study we have investigated the determination of proper time step in molecular dynamics simulation. Since the molecular dynamics is mathematically related to nonlinear dynamics,¹¹ the analysis of eigenvalues is used to explain the relationship between the time step and dynamics.¹²

The tracings of H₂ and CO₂ molecular dynamics simula-

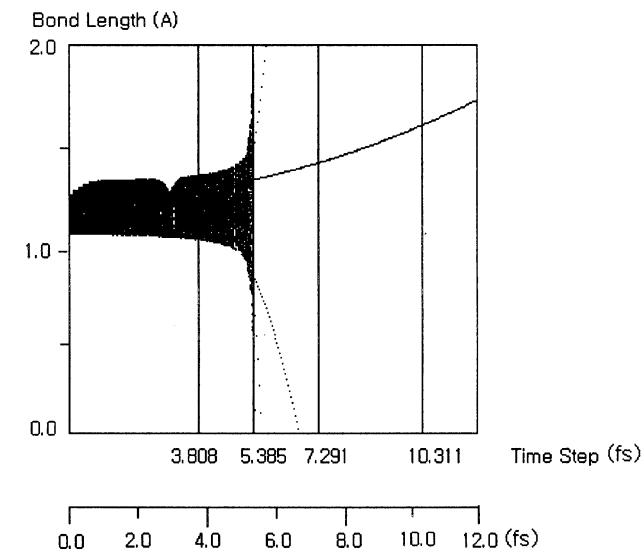


Figure 5. MD orbit diagram of CO₂ molecule.

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Although this results were derived for most simple cases of hydrogen and carbon dioxide, we could quantitatively explain why improperly large time step destroyed the molecular dynamics. From this study we could set the guide line of the proper time step for stable dynamics simulation in molecular modeling software.

Acknowledgment. This research was supported by the

Chung-Ang university research grants in 1998.

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