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Quantum molecular dynamics study of light-to-heat absorption mechanism in atomic systems

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Abstract—The quantum molecular dynamics method is applied to understand the fundamental mechanism of light-to-heat conversion in atomic systems under light irradiation. The light energy governs the time history of kinetic energy of the atomic system under light irradiation. Under infra-red light irradiation, the atomic fragments made by light irradiation have translational velocities parallel to the direction of light fluctuation. Under light irradiation at electron energy levels, the fragments tend to be isolated atoms having random translational velocities. The light electric field interacts with the atomic system through different terms of the molecular dynamics equations in the thermal conversion processes of light in the visible to infra-red range. Under infra-red light irradiation, the light interaction comes through fluctuations of effective dipole moment, whereas light irradiation at electron energy levels, it comes from changes in the potential energy between atoms associated with the electronic excitation. © 1997 Elsevier Science Ltd.

1. INTRODUCTION

The progress of laser technology has made it necessary to understand the light interaction processes with matters in non-equilibrium state. Although laser manufacturing of materials has been applied widely, it is hardly explained theoretically how the light energy at electron excitation levels is converted into atomic thermal energies. On the other hand, according to the progress of computational hardware the process of light interaction with matters has been studied recently by solving the Schrödinger equation. Metiu *et al.* calculated the photo-dissociation process of two atomic molecules [1]; Heather, H_2^+ [2] in intense light field; Doublet *et al.*, H_2O [3]; Garcia-Vele, Ar-HCl [4]; Guo, HBr on LiF (001) surface [5] and NH_3 on metal surface [6]; Atabek *et al.*, H_2^+ [7]; H. Zhang *et al.*, H_2O_2 [8]. Generally the time-dependent Schrödinger equation is solved by propagating the initial wave functions with the split operator method [9–12] with the FFT technique developed by Feit *et al.* [13]. These time-dependent approaches are usually applied to simple atomic or molecular system to obtain the dynamic features under light irradiation. From an engineering point of view, it is necessary to develop a numerical method which can be applied to large

systems under light irradiation at wide energy levels.

On the other hand, the molecular dynamics method (MD) has been developed and applied to molecular systems for understanding the fundamental mechanism of thermal problems such as condensation or heat conduction [14]. Since light absorption is related to changes in rotational, vibrational or electronic state of molecules, it is possible to apply the molecular dynamics method to the thermal process of light absorption. The potential energy used in the MD method should be changed by light absorption during the processes. This makes it difficult to analyze such phenomena with the MD method [15].

In the previous report [16], the quantum molecular dynamics method (QMD) is introduced to the calculation of light interaction problem with matters and it is applied to a system of two metallic atoms. The results show that the thermal processes of light irradiation are mainly attributed to the change in the atomic dipole moment under infra-red light irradiation and to the change in the atomic potential energy under light irradiation at electronic excitation levels. In the present study, the quantum molecular dynamics method is applied to systems of seven and thirteen atoms under light irradiation and the light-

NOMENCLATURE

d_{ij}^k, b_{ij}^k	numerical function at grid point (i, j)	β_{loc}	dipole moment of localized free electrons
e	unit electric charge	β_{deloc}	dipole moment of delocalized free electrons
$e \cdot Z$	electric charge of a nucleus	Δt	time step
\mathbf{E}	light electric field	ε	energy parameter of the ground Lennard-Jones potential energy
E_0	magnitude of light electric field	θ	light direction angle
H	Hamiltonian	σ	scale parameter of the ground Lennard-Jones potential energy
K	kinetic energy	ρ_k	density of a free electron (k)
L_0	length of calculation area	φ	light phase angle
m_e	mass of an electron	ψ_{fe}^k	wave function of a free electron (k)
M_i	mass of an ion	Ψ	wave function of the system
N	the number of grid points	ω	light angular frequency.
q_{mn}	effective dipole moment of ions		
\mathbf{r}^j	coordinates of an electron (j)		
\mathbf{R}_i	coordinates of ions		
\mathbf{R}^k	coordinates of nuclei (k)		
R_{mn}	distance between ion (m) and ion (n) with light irradiation		
R_{mn0}	distance between ion (m) and ion (n) without light irradiation		
S	deformation index		
U	potential energy		
U_{local}^{i-i}	potential energy between ions with localized free electrons		
U_{deloc}^{i-i}	potential energy between ions with delocalized free electrons.		
Greek symbols			
α	overlap parameter of potential energy between ions		
β	effective dipole moment of free electrons with light irradiation		
		Superscripts/subscripts	
		A-B	between A and B
		be	bound electrons
		deloc	delocalized free electrons
		e	electrons
		fe	free electrons
		i	ions
		k	number (k) of an ion or an electron
		light	light effects
		local, loc	localized free electrons
		n	nuclei
		*	dimensionless value
		'	excited state.

to-heat conversion mechanism is studied by observing the atomic motions associated with changes in system and light parameters.

2. NUMERICAL METHOD

When an atomic system is composed of atomic nuclei (n) and electrons (e), the time dependent Schrödinger equation of this system under light irradiation can be expressed as follows:

$$i\hbar \frac{\partial}{\partial t} \Psi = H\Psi = (-K + U + U_{light})\Psi. \quad (1)$$

The Hamiltonian can be written as,

$$H = -K_n - K_e + U_{n-n} + U_{n-e} + U_{e-e} + U_{light-n} + U_{light-e} \quad (2)$$

where $U_{light-n}$ and $U_{light-e}$ are the potential of nuclei and electrons due to the electric field of light given by,

$$U_{light-n} = \sum_k e \cdot Z_k \cdot \mathbf{R}^k \cdot \mathbf{E} \quad (3)$$

$$U_{light-e} = \sum_j (-e) \cdot \mathbf{r}^j \cdot \mathbf{E}. \quad (4)$$

The electric charge of nuclei, coordinates of nuclei and coordinates of electrons are denoted by $e \cdot Z$, \mathbf{R} and \mathbf{r} , respectively. The magnetic field of light is ignored in comparison with the electric field. The light electric field \mathbf{E} is expressed as,

$$\mathbf{E} = E_0 \cdot \cos \theta \cdot \cos(\omega t + \varphi) \quad (5)$$

where θ , ω and φ are the direction angle, angular frequency and the phase angle of light, respectively.

Electrons can be classified in two kinds of the behavior to nuclei; bound electrons (be) and free electrons (fe). The Hamiltonian is then written as

$$H = -K_n - K_{be} - K_{fe} + U_{n-n} + U_{n-be} + U_{n-fe} + U_{be-be} + U_{be-fe} + U_{fe-fe}$$

$$+ U_{\text{light}-n} + U_{\text{light}-\text{be}} + U_{\text{light}-\text{fe}}. \quad (6)$$

By considering the motion of nuclei and electrons, the wave function can be assumed to be separable in a form of

$$\Psi = \Psi_n \cdot \Psi_{\text{be}} \cdot \Psi_{\text{fe}} \quad (7)$$

although Ψ_{be} and Ψ_{fe} are functions of \mathbf{R} and \mathbf{r} . Nucleus motions are much related to the average state of Ψ_{be} and Ψ_{fe} . Free electrons can be considered as the electrons uncoupled from the bound ones due to their weakly coupling potential energy to the nuclei so that the bound electrons and their nucleus can be considered to form an ion (i). The wave function can be then assumed as:

$$\Psi = \Psi_i \cdot \Psi_{\text{fe}}. \quad (8)$$

The governing equations are then reduced to

$$i\hbar \frac{\partial}{\partial t} \Psi_{\text{fe}} = \{-K_{\text{fe}} + U_{\text{fe}-\text{fe}} + U_{i-\text{fe}} + U_{\text{light}-\text{fe}}\} \Psi_{\text{fe}} \quad (9)$$

$$i\hbar \frac{\partial}{\partial t} \Psi_i = \{-K_i + U_{i-i} + U_{i-\text{fe}} + U_{\text{light}-i}\} \Psi_i \quad (10)$$

where

$$U_{\text{light}-\text{fe}} = \sum_k e \cdot (\mathbf{R}^k - \mathbf{r}^k) \cdot \mathbf{E}. \quad (11)$$

The Newton's equation can be obtained for ions by ignoring the distribution of Ψ_i and assuming $\hbar \rightarrow 0$.

$$M_i \frac{\partial^2 \mathbf{R}_i}{\partial t^2} = - \frac{\partial}{\partial \mathbf{R}_i} \{ \langle \Psi_{\text{fe}} | U_{i-i} | \Psi_{\text{fe}} \rangle + \langle \Psi_{\text{fe}} | U_{i-\text{fe}} | \Psi_{\text{fe}} \rangle + \langle \Psi_{\text{fe}} | U_{\text{light}-i} | \Psi_{\text{fe}} \rangle \}. \quad (12)$$

The term $U_{\text{light}-i}$ includes the light interaction with the bound electrons, that is, the effect of polarization of ions. It can be assumed that the polarization induces an effective dipole moment so as to be expressed as

$$\langle \Psi_{\text{fe}} | U_{\text{light}-i} | \Psi_{\text{fe}} \rangle = \sum_{m,n} q_{mn} \cdot (\mathbf{R}_{mn} - \mathbf{R}_{mn0}) \cdot \mathbf{E} \quad (13)$$

where R_{mn} and R_{mn0} are the distance between ion (m) and ion (n) with and without light irradiation, respectively. q_{mn} is the magnitude of the induced dipole moment with light irradiation.

The wave function of free electrons can be obtained by solving the Schrödinger equation equation (9). With this Ψ_{fe} , the Newton's equation, equation (12), can be solved to obtain the motion of ions. With the assumption that the free electrons are not strongly related with each other, the Schrödinger equation can be separated into the time dependent Schrödinger equations for each electron, ψ_{fe}^k .

Other contributions of the light irradiation to the molecular motion are changes in the ionic potentials $U_{i-\text{fe}}$ and U_{i-i} , which include electronic interactions of the bound electrons. The potential between ion and free electron can be expressed as

$$\langle \Psi_{\text{fe}} | U_{i-\text{fe}} | \Psi_{\text{fe}} \rangle = \sum_k \rho_k \cdot U_{i-\text{fe}}^k \quad (14)$$

where

$$\rho_k = \langle \psi_{\text{fe}}^k | \psi_{\text{fe}}^k \rangle. \quad (15)$$

The electronic states of the bound electrons are changeable with the free electron field, it leads to modify the potential between ions. Light irradiation makes the electronic state of free electrons excite, say, from state 1 to 2. At state 1, free electrons are at the ground state localized around ions, and at state 2 they are sufficiently distributed and delocalized by light absorption. Corresponding to these states 1 and 2, denote the potential between ions as U_{local}^{i-i} and U_{deloc}^{i-i} , respectively. The potential energy during excitation takes intermediate values between them. The intermediate may be assumed to take a form of linear summation as,

$$\langle \Psi_{\text{fe}} | U_{i-i} | \Psi_{\text{fe}} \rangle = \alpha \cdot U_{\text{local}}^{i-i} + (1-\alpha) \cdot U_{\text{deloc}}^{i-i} \equiv U_{i-i}. \quad (16)$$

In the present study, U_{local}^{i-i} and U_{deloc}^{i-i} are expressed by the Lennard-Jones potential as:

$$U_{\text{local},mn}^{i-i} = 4\epsilon \left\{ \left(\frac{\sigma}{R_{mn}} \right)^{12} - \left(\frac{\sigma}{R_{mn}} \right)^6 \right\} \quad (17)$$

$$U_{\text{deloc},mn}^{i-i} = 4\epsilon' \left\{ \left(\frac{\sigma'}{R_{mn}} \right)^{12} - \left(\frac{\sigma'}{R_{mn}} \right)^6 \right\}. \quad (18)$$

The parameter α can be a function of positions of free electrons which are related to the effective dipole moment of free electrons β ;

$$\beta = \sum_{k=i,j} \sum_{\text{allspace}} e \cdot \rho(\mathbf{r}_{\text{fe}}^k) |\mathbf{R}_i^k - \mathbf{r}_{\text{fe}}^k|. \quad (19)$$

If the parameter α is assumed to be proportional to β , α is given by:

$$\alpha = \{\beta_{\text{deloc}} - \beta\} / \{\beta_{\text{deloc}} - \beta_{\text{loc}}\}. \quad (20)$$

The potential energy between ions is U_{local}^{i-i} at $\beta = \beta_{\text{loc}}$ and U_{deloc}^{i-i} at $\beta = \beta_{\text{deloc}}$.

The Schrödinger equation for a free electron (k) and the Newton's equation for ions (m) and are finally described as,

$$i\hbar \frac{\partial \psi_{\text{fe}}^k}{\partial t} = \{-K_{\text{fe}}^k + U_{\text{fe}-\text{fe}}^k + U_{i-\text{fe}}^k + e \cdot \mathbf{E} \cdot (\mathbf{R}^k - \mathbf{r}^k)\} \psi_{\text{fe}}^k \quad (k = 1, 2, \dots) \quad (21)$$

$$M_i \frac{\partial^2 \mathbf{R}_i^m}{\partial t^2} = - \frac{\partial}{\partial \mathbf{R}_i^m} \left\{ U_{i-i}^m + \sum_k \rho_k \cdot U_{i-\text{fe}}^{m,k} + \sum_{m,n} q_{mn} \cdot \mathbf{E} \cdot (\mathbf{R}_{mn} - \mathbf{R}_{mn0}) \right\} \quad (m = 1, 2, \dots) \quad (22)$$

where the Coulomb type potential energy is used for $U_{i-\text{fe}}$ and $U_{\text{fe}-\text{fe}}$. The system parameters included in

Table 1. System parameters in the calculation

System parameters	Non-dimensional	Standard value
Energy parameter of the excited Lennard–Jones potential ε'	ε^*	8.125×10^{-20} (J)
Scale parameter of the excited Lennard–Jones potential σ'	σ^*	4.555×10^{-10} (m)
Dipole coefficient of bound electrons of ion q_{mn}	q^*	3.7×10^{-18} (C)
Dipole coefficient of ground free electron field β_{loc}	β_{loc}^*	$1.8 \times 10^{-10} \times e$ (C·m)
Dipole coefficient of excited free electron field β_{deloc}	β_{deloc}^*	$17 \times 10^{-10} \times e$ (C·m)

these equations and used in the present study are listed in Table 1. With the standard values, the system parameters are non-dimensionalized as (ε^* , σ^* , q^* , β_{loc}^* , β_{deloc}^*).

Equation (21) is simply written as :

$$i\hbar \frac{\partial \psi_{fe}}{\partial t} = (K + U)\psi_{fe} \quad (23)$$

where the kinetic operator is

$$K = -\frac{\hbar^2}{2m_e} \nabla^2. \quad (24)$$

The solution of equation (23) becomes a complex function of the time t and coordinates \mathbf{r} . The problems of solving the equation are the time integration and the second-order space derivatives of this equation. As for the time integration of equation (23), the split operator method [13] is employed in the present study as :

$$\begin{aligned} \psi_{fe}(t + \Delta t) = & \exp\left(\frac{i\hbar\Delta t}{4m_e} \nabla^2\right) \exp\left(\frac{-i\Delta t U}{\hbar}\right) \\ & \times \exp\left(\frac{i\hbar\Delta t}{4m_e} \nabla^2\right) \psi_{fe}(t). \end{aligned} \quad (25)$$

In order to keep sufficient precision of the second-order derivatives on the fixed space grid points, it is necessary to prepare very fine grid points which cost long calculation time. In most cases, the function expansion method is employed with Fourier expansion [13] or Chebyshev polynomials. The Fourier expansion method is applied in the present study. The Fourier expansion of a two-dimensional wave function can be written as

$$\begin{aligned} \Psi(x, y, t) = & \sum_{m=-N/2+1}^{N/2} \sum_{n=-N/2+1}^{N/2} a_{mn}(t) \\ & \cdot \exp\left[\frac{2\pi i}{L_0}(mx + ny)\right] \end{aligned} \quad (26)$$

where N , $a_{mn}(t)$ and L_0 are the number of grid points, the coefficients in Fourier space and the length of calculation area, respectively ($N = 32$, $L_0 = 31 \text{ \AA}$). The wave function of free electrons is represented as complex numerical values at two dimensional grid points (i, j) in a form of

$$\psi_{fe,ij}^k = a_{ij}^k + ib_{ij}^k \quad (k = 1, 2, \dots). \quad (27)$$

The velocity and position of ions are calculated by the molecular dynamics method with using the central difference method [17].

3. RESULTS AND DISCUSSIONS

In the previous report [16], the quantum molecular dynamics method was applied to two-atom systems under light irradiation. In the present study, the QMD method is applied to atomic systems of seven and 13 atoms shown in Fig. 1. Light is irradiated to the system in the direction as shown by the arrow. In Section 3.1 light irradiation effects on the atomic motion are discussed from the point of the atomic deformation and the time history of kinetic energy. In Section 3.2 light to heat conversion mechanism is studied by changing the parameters of the system and light irradiation in order to know which terms of the Newton's equation govern the process predominantly.

3.1. Light irradiation effects on atomic motions

The effects of light irradiation on atomic motions are discussed in this section by examining the atomic deformation and time history of the kinetic energy under light irradiation. Figure 2 shows ion positions and electric charge density distribution at 100, 200 and 300 fs under light irradiation for a seven-atom

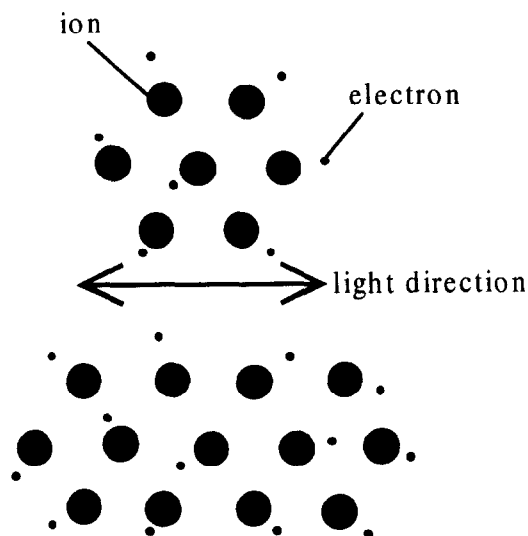


Fig. 1. Configuration of ions and free electrons.

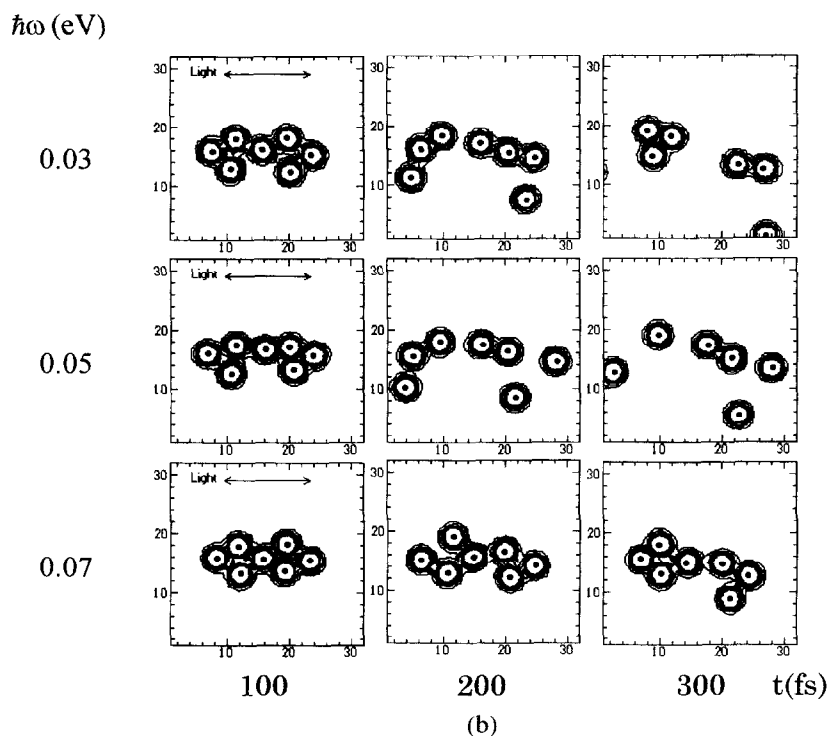
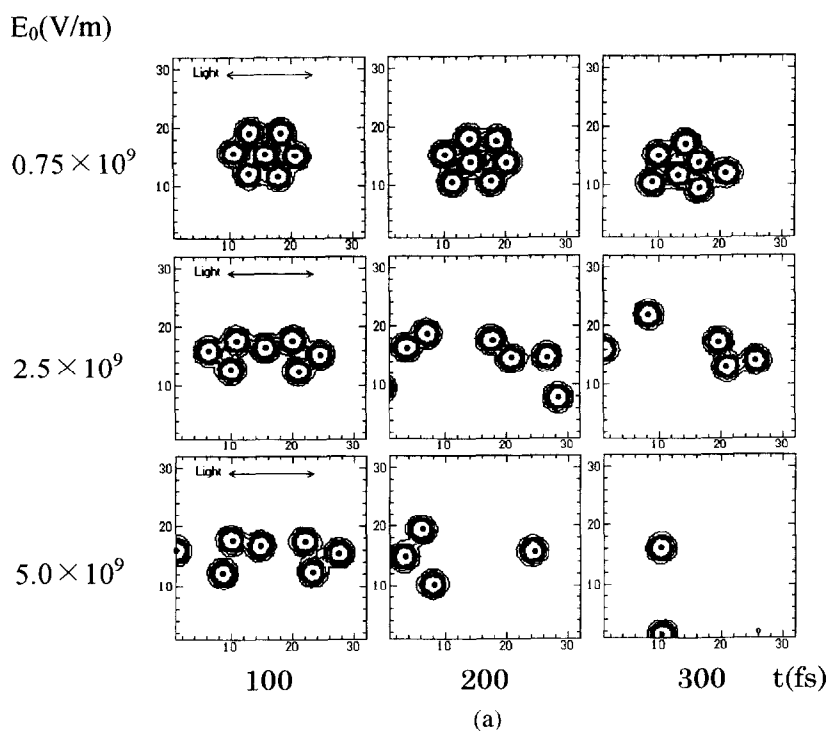


Fig. 2. (a) Atomic deformation under light irradiation: effect of light electric field (seven atom system); (b) atomic deformation under light irradiation: effect of light energy (seven atom system); (c) atomic deformation under light irradiation: effect of light energy (seven atom system).

system. The light energy is 0.03 eV which corresponds to an infra-red light energy level and the light electric fields are 0.75×10^9 , 2.5×10^9 and 5.0×10^9 V m⁻¹,

which are comparable with practical laser outputs. Light parameters are written in Table 2. Dimensionless parameters of the system (e^* , σ^* , q^* ,

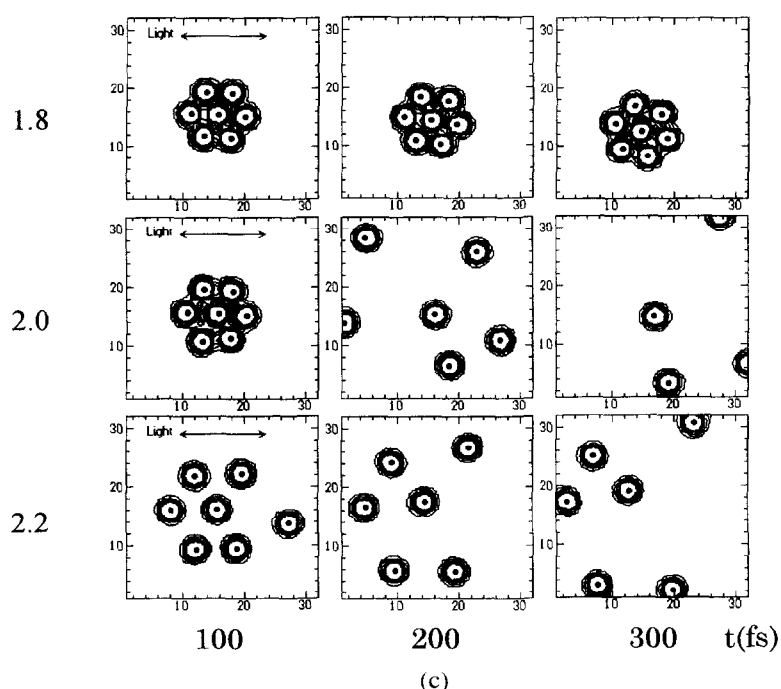


Fig. 2—continued.

Table 2. Energy and electric field of light

	Light energy (eV)	Light electric field (10^9 V m^{-1})
Case 1 [Fig. 2(a)]	0.03	0.75
Case 2 [Fig. 2(a)]	0.03	2.5
Case 3 [Fig. 2(a)]	0.03	5.0
Case 4 [Fig. 2(b)]	0.03	2.0
Case 5 [Fig. 2(b)]	0.05	2.0
Case 6 [Fig. 2(b)]	0.07	2.0
Case 7 [Fig. 2(c)]	1.8	2.5
Case 8 [Fig. 2(c)]	2.0	2.5
Case 9 [Fig. 2(c)]	2.2	2.5

β_{loc}^* , β_{deloc}^* are (0.5, 2.0, 1.0, 1.0, 1.0), respectively. The cases with different system parameters are calculated, but the results are not shown here.

Figure 2(a) shows that the atomic system has been deformed to the arrow direction which represents the light direction. In the cases of 2.5×10^9 and $5.0 \times 10^9 \text{ V m}^{-1}$, the system is deformed and finally dissociated into atoms or atomic clusters. The fragments made by light irradiation tend to have a translational velocity parallel to the light direction. The characteristic time of these deformation depends on the light electric field, becoming short as the electric field is large. In Fig. 2(b), the light is irradiated with the energy of 0.03, 0.05 and 0.07 eV and the light electric field of $2.0 \times 10^9 \text{ V m}^{-1}$. In the case of 0.03 eV, the system is dissociated into two atoms, a dimer and a trimer at 300 fs. In the case of 0.05 eV, the system is dissociated into five atoms and a dimer. In the case of 0.07 eV, the seven-

atom system is deformed, but not dissociated by light irradiation within 300 fs. The natural frequency of the inter atomic vibrational motion corresponds to the light energy from 0.01 to 0.03 eV. This will be concerned with the efficiency of light-to-heat conversion. Figure 2(c) shows the cases of high light energy of 1.8, 2.0 and 2.2 eV with $2.5 \times 10^9 \text{ V m}^{-1}$. In the case of 1.8 eV the system deformation is not observed within 300 fs. In the cases of 2.0 and 2.2 eV the system is decomposed into isolated atoms that have random translational velocities parallel to the light direction. The atomic translational velocities are also different in both cases. It indicates that the light energy also affects the magnitude of translational velocity of the fragments.

Figure 3 shows the atomic configuration to thirteen atom system at 30, 60 and 90 fs under various light irradiation. Figure 4 is the time history of kinetic energy of the system under light irradiation shown in Table 3. System parameters (ϵ^* , σ^* , q^* , β_{loc}^* , β_{deloc}^*) are chosen as (0.5, 1.2, 1.0, 1.0, 0.18) because the potential energy with excited free electron is assumed to have a smaller ϵ and larger σ than the ground potential has. In the case of 0.01 eV the deformation of the system is not observed within 90 fs. In the case of 0.03 eV, the system is deformed and dissociated. In the case of 0.1 eV, the system deformation is observed and an atom is separated to form a (1+12) atom system. In the case of 0.3 eV, the deformation of the system is not observed. In the case of 2.0 eV, the system is dissociated into atoms and atomic clusters that have random translational velocities parallel to the light direction. The relationship between the light energy

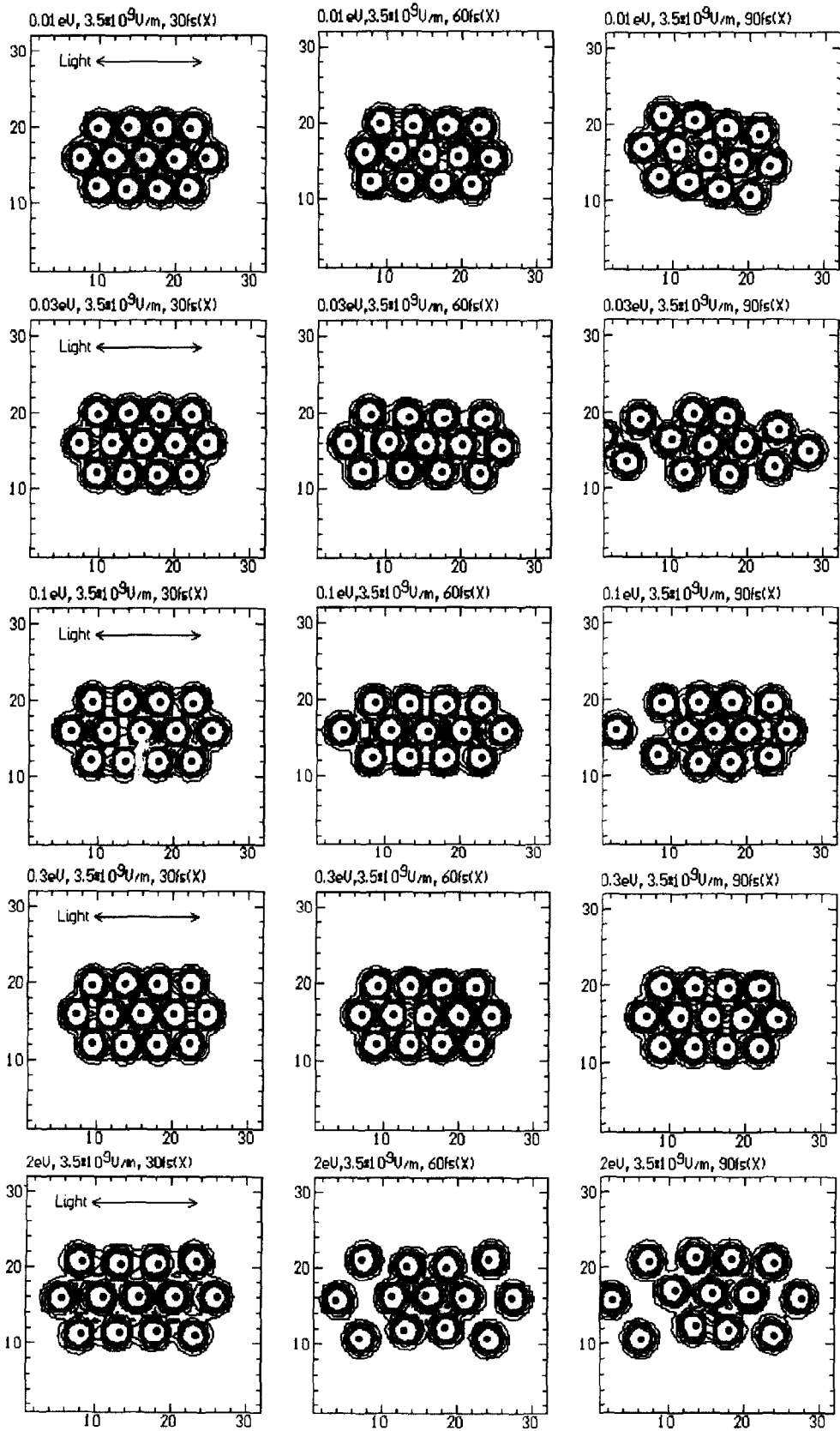


Fig. 3. Atomic deformation under light irradiation : effects of light energy (13 atom system).

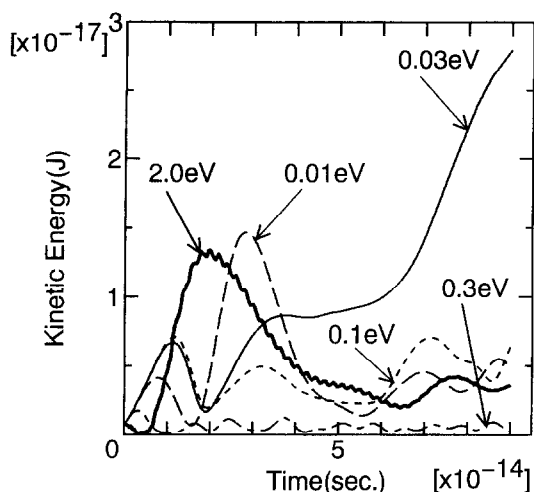


Fig. 4. Time history of the kinetic energy: effects of light energy (13 atom system).

Table 3. Energy and electric field of light

	Light energy (eV)	Light electric field (10^9 V m^{-1})
Case 1	0.01	3.5
Case 2	0.03	3.5
Case 3	0.1	3.5
Case 4	0.3	3.5
Case 5	2.0	3.5

and fragments motion is similar to that of the seven atom systems in Fig. 2. The time history of the vibrational kinetic energy shows different features depending on the light energy. In the cases of 0.01, 0.03, 0.1 and 2.0 eV, fluctuations are observed in the time history of kinetic energy although the case of 0.3 eV has little fluctuations. However, the kinetic energy itself is increased intensively only when the light energy is 0.03 eV. These characteristics of the atomic deformation and the time history of kinetic energy should be changed with different system parameters. In a present study the system deformation is associated with increasing the kinetic energy under light irradiation of 0.03 eV and without any increase of the kinetic energy under 2.0 eV irradiation.

3.2. Light-to-heat conversion mechanism

In Section 3.1, effects of light irradiation on the atomic configurations and motions are shown in various cases of light and system parameters. These features will be related with the processes from light absorption to heat conversion. In this section, the mechanism of light-to-heat absorption is studied by calculating the kinetic energy (K) and the deformation index (S) under various conditions of light irradiation and atomic system. The deformation index (S) is defined as,

$$S = \sum_{i=1}^{13} |\mathbf{R}_i - \mathbf{R}_0|/13 \quad (28)$$

where \mathbf{R}_i is the coordinates of an ion and \mathbf{R}_0 is the center of mass of the system. This value represents the magnitude of deformation of the atomic system and it is about 5.8×10^{-10} m at the stable state of the atomic system calculated in the present study. The calculation system is a 13 atom system as shown in Fig. 1. Light and system parameters used in this section are written in Tables 3 and 4, respectively.

Figure 5 shows the effects of light parameters on the deformation index (S) and kinetic energy (K) at 90 fs after light irradiation. In the case of the light electric field of $5.0 \times 10^9 \text{ V m}^{-1}$, both deformation index and kinetic energy become larger than those of $3.5 \times 10^9 \text{ V m}^{-1}$ at almost all light energies. Under irradiation of $3.5 \times 10^9 \text{ V m}^{-1}$, the light irradiation induces the atomic motion effectively at light energies of 0.03 and 2.0 eV and less effectively at light energies between these values. The figure shows that system deformation is resulted in most predominantly under light irradiation of 2.0 eV and $5 \times 10^9 \text{ V m}^{-1}$.

In Fig. 6, effects of the effective dipole moment (q^*) are shown on the deformation index and the kinetic energy. Decreasing the magnitude of the dipole moment leads to decrease in the light interaction except at light energy of 2.0 eV at which light irradiation induces the electronic excitation of atoms. Figure 7 shows the effects of system parameters σ^* and β_{deloc}^* on the deformation index (S) and kinetic energy (K). It is seen from the figure that these system parameters affect the atomic motion only when the light energy is 2.0 eV. The system parameter q^* is related with the third term of equation (22) and σ^* and β_{deloc}^* with the first term of equation (22). These results definitely explain that the light electric field interacts with the atomic system through the first term of equation (22) under the light irradiation at energy levels of electronic excitation and through the third term under infra-red light irradiation. The third term has direct relationship with the direction of the light electric field. In Section 3.1, there are some differences in the atomic deformation under light irradiation at infra-red energy levels and that at electron energy

Table 4. System parameters

	ε^*	σ^*	q^*	β_{loc}^*	β_{deloc}^*
Case 1 (Fig. 5)	0.5	1.2	1.0	1.0	0.18
Case 2 (Fig. 6)	0.5	1.2	0.1	1.0	0.18
Case 3 (Fig. 7)	1.0	1.4	1.0	1.0	0.18
Case 4 (Fig. 7)	1.0	1.3	1.0	1.0	0.18
Case 5 (Fig. 7)	1.0	1.2	1.0	1.0	0.18
Case 6 (Fig. 7)	0.5	1.2	1.0	1.0	0.176
Case 7 (Fig. 7)	0.5	1.2	1.0	1.0	0.353
Case 8 (Fig. 7)	0.5	1.2	1.0	1.0	0.529
Case 9 (Fig. 7)	0.5	1.2	1.0	1.0	1.0

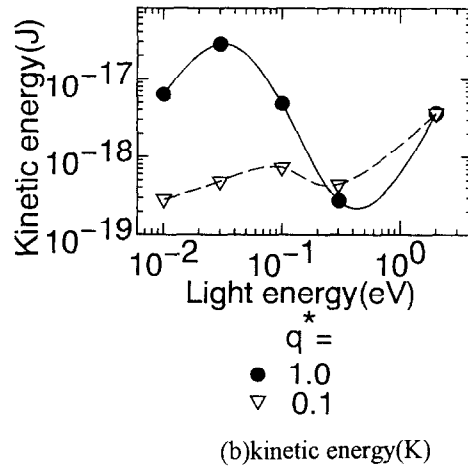
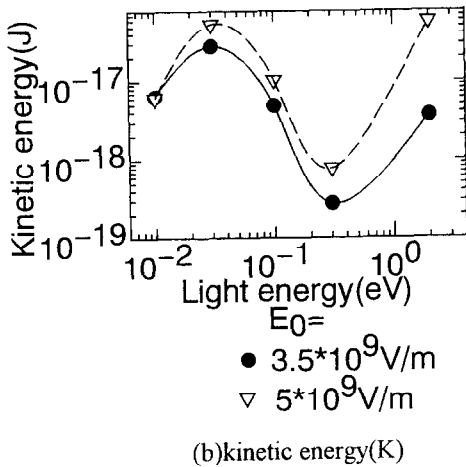
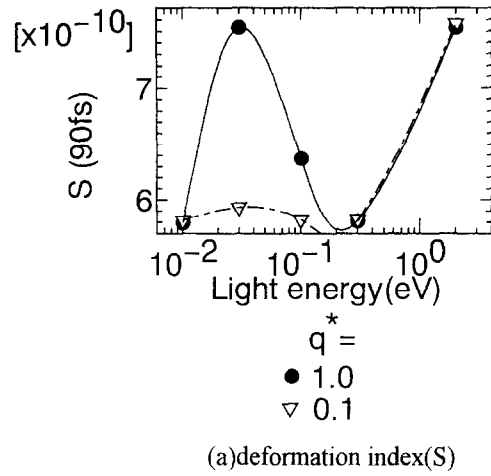
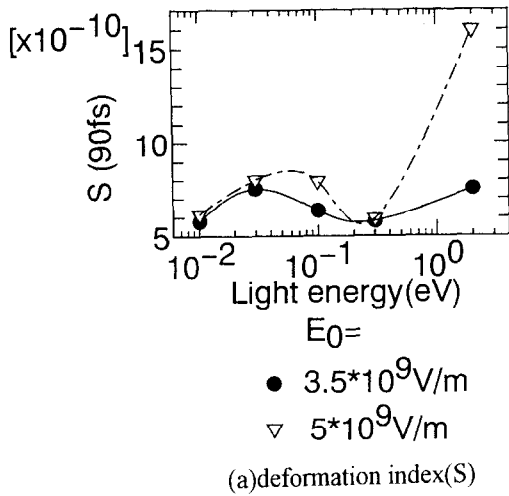


Fig. 5. Effects of light parameters on deformation index (S) and kinetic energy (K): (a) deformation index (S); (b) kinetic energy (K).

Fig. 6. Effects of system parameter (q^*) on deformation index (S) and kinetic energy (K): (a) deformation index (S); (b) kinetic energy (K).

levels. This is because light absorption at electron energy levels changes the potential between ions through the first term in equation (22) and this potential change hardly depends on the light direction, and even if the atomic system is decomposed into atomic clusters such as dimers and trimers, the decomposed cluster has a large possibility to absorb the light energy regardless of its directional configuration with respect to the light direction. It means that the light irradiation keeps continuous effects on the atomic motion after the decomposition, but light absorption at infra-red energy levels is caused by the effective dipole fluctuation with the light electric field through the third term in equation (22). It has the largest effects on the atomic system when the system axis is parallel to the light direction. When the atomic system is dissociated into atomic dimers or smaller clusters to get rotational motions, the rotational motions may reduce the possibility of light absorption. This is the reason why the fragments of atomic cluster tends to

have parallel translational velocities to the light direction. Differences of the mechanism of light absorption to heat conversion due to the light energy cause the differences in the thermal processes of the atomic system, the time history of kinetic energy and system deformation.

4. CONCLUSION

The quantum molecular dynamics method is developed to analyze the fundamental mechanism of light-to-heat conversion mechanism. The numerical method developed in the present study can be applicable to the atomic system under light irradiation from infra-red light to visible light. Characteristic differences in the kinetic energy and deformation of the system are observed under various light irradiation. Under infra-red light irradiation, the atomic fragments made by light irradiation have parallel trans-

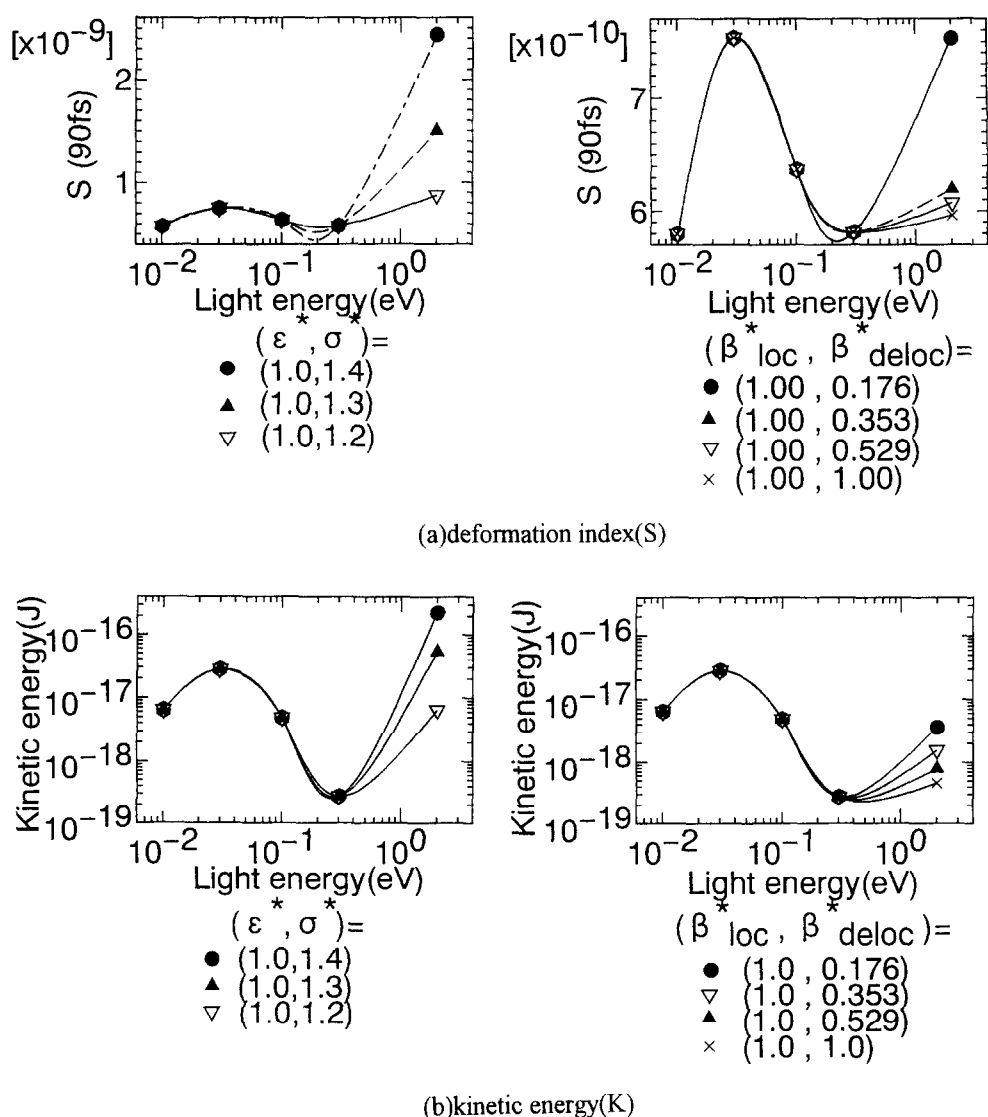


Fig. 7. Effects of system parameters (ϵ^* , σ^* , β_{loc}^* , β_{deloc}^*) on deformation index (S) and kinetic energy (K): (a) deformation index (S); (b) kinetic energy (K).

lational velocities. Under light irradiation at electron energy levels, the fragments tend to be isolated atoms having random translational velocities to the light direction. These differences are caused by the difference of the light-to-heat absorption mechanism depending on the light energy. Under infra-red light irradiation, the light interacts with the atomic system through an effective dipole moment fluctuation, and under the light irradiation at electron energy levels, it acts through changes in the potential energy between ions due to electronic excitation.

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