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Quantum molecular dynamics study on light-to-heat absorption mechanism: two metallic atom system

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Abstract—For understanding the fundamental mechanism of light absorption to be converted to the thermal energy, the atomic motions of matters are studied with the quantum molecular dynamics method. As a fundamental atomic system, two metallic atoms are considered to be irradiated with light changing the frequency and intensity. Under infra-red light irradiation, increase in the kinetic energy and atomic dissociation are attributed mainly from the dipole moment fluctuations of ions. Under the irradiation of light at electron energy levels, change in the kinetic energy and dissociation result from the change in the potential energy between particles due to electron excitation. © 1997 Elsevier Science Ltd.

1. INTRODUCTION

Radiative heat transfer is one of the fundamental processes of heat transfer and it is usually dealt with as a problem of thermal energy transfer. The progress of optical technology, such as laser processing, laser control of material manufacturing and laser cooling, has made it necessary to analyze and consider the non-equilibrium phenomena related to the light interaction with matter within very short time (fs, ps, ns). Basically such phenomena are problems of interaction between charged particles (electron, nuclei) and the electric field of light. These interactions have quantum aspects so that the processes can be understood from the quantum mechanical point of view even for the problems at the thermal energy levels.

The interaction between light and matter concerns engineering problems such as laser dissociation and ablation; which type of laser (what range of wave length and what magnitude of output) with how long time irradiation can dissociate the condensate into atoms or which type of laser under what conditions is the best for making the solid material into the vapor state. In these problems effects of the light or laser irradiation are always estimated with some coefficient introduced by experiments because the light energy is viewed only as a heat source without any consideration into the quantum and molecular behavior of light interaction with matter. On the other hand, the strict treatment of its quantum dynamics is not always useful for these problems because of computational difficulties associated with large number of electrons to be involved. In the process of light-to-heat absorption, the light absorption is dominated by the quantum behavior of light interaction with matter whereas its conversion to heat or thermal energy is related to the kinetic motion of atoms or molecules. This consideration, thus, makes it possible to treat the process with the quantum molecular dynamics so that the quantum dynamics analyzes the electronic states under light irradiation or electric field of light and the molecular dynamics explains the atomic or molecular motion under the potential associated with the electronic states.

In the present study, the atomic system is assumed simply to consist of ions and electrons where the ion has a closed shell with a small amount of ionization energy. The kinetic motion of ions can be predicted by the molecular dynamics method and the wave function of electrons can be obtained by the time-dependent Schrödinger equation [1–3]. The light can be considered basically as an electromagnetic field. The photon energy and energy density of light (or laser) are related to the frequency and magnitude of the light electric fields, respectively. The light electric field concerns the potential terms of the Newton's equation of ions and the Schrödinger equation of electrons. With this quantum molecular dynamics method, the relationships between light irradiation, kinetic energy change and atomic dissociation are discussed qualitatively in order to understand the fundamental mechanism of the interaction between light and matters.

2. NUMERICAL METHOD

In a system composed of electrons, ions and atoms, electron states should be treated quantum mechanically since the light absorption means changes in

NOMENCLATURE

e	unit electric charge
$\mathbf{E}$	light electric field
E_g	ground energy of valence electron
E_0	magnitude of light electric field
$\hbar$	Planck constant/2π
H	Hamiltonian
K	kinetic energy operator $[=(-\hbar^2/2M)\nabla^2]$
M^i	mass of ion
m_e	mass of electron
q	dipole moment of ions
R	distance between ions
R_c	cut off distance of potential between ion and electron
R_0	stable distance between ions
t	time
U	potential energy
U^e	potential energy of valence electrons
U^{e-e}	potential energy between electrons
U^{i-e}	potential energy between ion and valence electron
$U^{i-i}_{\rm local}$	potential energy between ions of localized valence electrons without light irradiation
$U^{i-i}_{\rm deloc}$	potential energy between ions of delocalized valence electrons with light irradiation
$U^{\rm light}$	potential energy of light interaction
$U^{\rm light}_{\rm ion}$	potential energy of ions by light irradiation
$U^{\rm light}_{\rm elec}$	potential energy of valence electrons by light irradiation
X^i	ion coordinate
x^e	electron coordinate.

Greek symbols

α	overlap parameter of potential energy between ions
β	dipole moment of valence electrons with light irradiation
$\beta_{\rm loc}$	dipole moment of localized valence electrons
$\beta_{\rm deloc}$	dipole moment of delocalized valence electrons
Δt	time step
ε	Lennard–Jones potential parameter
ε^*	dimensionless Lennard–Jones potential parameter
θ	directional angle of light
ρ	electron charge density
σ	Lennard–Jones potential parameter
σ^*	dimensionless Lennard–Jones potential parameter
$\sigma(\omega)$	cross-section spectra of light absorption
φ	phase angle of light
Ψ	wave function of the system
Ψ^e	wave function of valence electrons
Ψ^i	wave function of ions
ω	angular frequency of light.

the quantum number of the system. When light or laser output is strong enough, electrons and atoms have a chance to be dissociated from the atom or molecule. The wave function of electronic states of the system can be calculated by the time-dependent Schrödinger equation. In these systems, however, the full quantum treatment of light interaction is too complicated even with a largest powerful computer to obtain a meaningful solution of the time-dependent Schrödinger equations. At thermal energy levels, inter-molecular quantum states can be assumed to be continuous so that the motions of ions can be solved with classical molecular dynamics. In the present study, for the purpose of the fundamental understanding of light-to-heat mechanism and for the development of the calculation methods of the phenomena, the atomic system of the matter interacting with light is assumed simply to consist of a valence electron and an ion composed of bound electrons and nuclei. For such an atomic system, the quantum states of the valence electron can be solved with the time dependent Schrödinger equation and the atomic motions with the molecular dynamics method.

The system consists of n ions(i) and n electrons(e). The time dependent Schrödinger equation of the system under light irradiation is written as

$$i\hbar\frac{\partial\Psi}{\partial t} = H\Psi = \{K+U+U^{\rm light}\}\Psi \tag{1}$$

where H, K and U are the Hamiltonian, kinetic operator ($=(-\hbar^2/2m)\nabla^2$) and potential energy, respectively. $U^{\rm light}$ is the potential of light interaction. Due to the difference in the mass and the characteristic time of particle motion, the wave function of the system Ψ is separable into the valence electrons Ψ^e and ions Ψ^i.

For ions, the Newton's equations can be obtained so far as the distributions of wave functions Ψ^i are completely ignored;

$$M^i\frac{{\rm d}^2\mathbf{X}^i}{{\rm d}t^2} = \frac{\partial}{\partial\mathbf{X}^i}\{\langle U^{i-e}\rangle_e + U^{i-i} + \langle U^{\rm light}\rangle_e\} \tag{2}$$

where

$$\langle U\rangle_e \equiv \langle\Psi^e|U|\Psi^e\rangle.$$

U^{i-e} and U^{i-i} are the potential energy between ion and electron and that between ions, respectively.

For the wave function of valence electrons Ψ^e, the following time-dependent Schrödinger equation is obtained;

$$i\hbar\frac{\partial\Psi^{e}}{\partial t} = \{K^{e}+U^{i-e}+U^{e-e}+\langle U^{\mathrm{light}}\rangle_{i}\}\Psi^{e} \tag{3}$$

where

$$\langle U\rangle_{i} \equiv \langle\Psi^{i}|U|\Psi^{i}\rangle.$$

U^{e-e} is the potential energy between electrons.

Since the magnetic effect of light can be ignored in comparison with the electric effect, the light electric field is expressed as,

$$\mathbf{E} = E_{0}\cdot\cos\theta\cdot\cos(\omega t+\varphi) \tag{4}$$

where E_0 is the magnitude of the electric field, θ is the directional angle and φ the phase angle.

The potential of light interaction in equations (2) and (3) has two contribution from ions and valence electrons;

$$U^{\mathrm{light}} = U^{\mathrm{light}}_{\mathrm{ion}}+U^{\mathrm{light}}_{\mathrm{elec}}. \tag{5}$$

Although the former should be described by full quantum dynamics for a couple of bound electrons and nuclei, it is assumed to be expressed in an effective dipole moment of ions q;

$$U^{\mathrm{light}}_{\mathrm{ion}} = q\cdot(\mathbf{R}-\mathbf{R}_{0})\cdot\mathbf{E} \tag{6}$$

where $\mathbf{R}$ and $\mathbf{R}_0$ are the distance between ions and stable distance between them. The latter is given by

$$U^{\mathrm{light}}_{\mathrm{elec}} = -e\cdot\left\{\sum_{n}(\mathbf{x}^{e}-\mathbf{X}^{i})\right\}\cdot\mathbf{E} \tag{7}$$

where $\mathbf{X}^i$ and $\mathbf{x}^e$ are ion and electron coordinates, respectively.

For the potentials between particles, the simplest and the most physically perspective form of Lennard–Jones and Coulomb type potential are assumed as follows.

$$U^{i-i} = 4\varepsilon\left\{\left(\frac{\sigma}{R^{i-i}}\right)^{12}-\left(\frac{\sigma}{R^{i-i}}\right)^{6}\right\}$$

$$U^{i-e} = -\frac{e^{2}}{4\pi\varepsilon_{0}R_{c}}(R_{c}\geqq R^{i-e}),$$

$$-\frac{e^{2}}{4\pi\varepsilon_{0}R^{i-e}}(R_{c}\leqq R^{i-e})$$

$$U^{e-e} = \frac{e^{2}}{4\pi\varepsilon_{0}R^{e-e}}. \tag{8}$$

In realistic meaning, U^{i-i} can be considered to be the summation of the Coulomb potential and L.J. potential. But to see the state of dissociation by potential energy change, only the L.J. potential is used for the potential between ions U^{i-i} in the present study. R_c is the cut off distance of U^{i-e} and 1.96 Å is used here. Equation (3) can be separated into two time dependent Schrödinger equations of electrons and wave functions of electrons are calculated by these equations. The potential energy U^{e-e} in equation (3) is calculated by the potential U^{e-e} in equation (8) and wave function of each electron.

The potential between ions U^{i-i} under light irradiation should be a complex function of positions of both valence electrons and ions to be obtained with the full quantum calculation. The states of electrons in ions should be, thus, changed by the valence electron distribution. In the present study, as the first approximation, this effect is taken into account in the following simple way. When the valence electrons are in ground states, the potential between ions is assumed to be U^{i-i}_{local}, that is, the Lennard–Jones type potential with two parameters. When the valence electrons are excited by light irradiation to have large kinetic energies to be delocalized, the potential is assumed to be U^{i-i}_{deloc} that is the Lennard–Jones type potential with different parameters from those of U^{i-i}_{local}. During the excitation process of light irradiation, the system can be considered to take a overlap state of these two states; that is

$$U^{i-i} = \alpha\cdot U^{i-i}_{\mathrm{local}}+(1-\alpha)\cdot U^{i-i}_{\mathrm{deloc}}. \tag{9}$$

Concerning the overlap parameter α, the realization of the states is assumed to be linearly proportional to the dipole moment of the valence electrons β;

$$\alpha = \{\beta_{\mathrm{deloc}}-\beta\}/\{\beta_{\mathrm{deloc}}-\beta_{\mathrm{loc}}\}$$

$$\beta = \sum_{i,j}\sum_{\mathrm{allspace}} e\cdot\rho(\mathbf{x}^{e})|\mathbf{X}^{i}-\mathbf{x}^{e}|, \tag{10}$$

where β_{loc} and β_{deloc} are β with localized electrons and β with delocalized electrons, respectively.

For light absorption, major parameters of light are light angular frequency ω and light electric field E_0 and those of the system, are dipole moment q and potential between ions U^{i-i} in excited states. There are four main parameters except for initial conditions of the system in this calculation as follows:

light

$$\omega, E_{0} \tag{11}$$

system

$$q, U^{i-i}.$$

The cross-section spectra of light absorption of valence electron can be defined as [1,2],

$$\sigma(\omega) \approx \omega\int_{0}^{\infty}\exp(i\omega t)C(t)\,\mathrm{d}t$$

$$C(t) = \exp(iE_{g}t)\langle\Psi^{e}(0)|\Psi^{e}(t)\rangle \tag{12}$$

where E_g is the ground energy of the system and ω the light angular frequency.

For the numerical calculation of equation (3), the split operator method [4] is employed as,

$$\Psi^{e}(t_0+\Delta t) = \exp\left(\frac{i\hbar\Delta t}{4m_e}\nabla^2\right)\exp(-i\Delta t U^{e}/\hbar) \times \exp\left(\frac{i\hbar\Delta t}{4m_e}\nabla^2\right)\Psi^{e}(t_0) \quad (13)$$

where U^e is the potential term in equation (3).

The wave function of valence electrons is evaluated at 32×32 grid points (point interval = 1 Å) and time-propagated with the time step of 10^{-17} second. The correlation or exchange energy between electrons is not considered here as the first approximation, although they play an important role in condensed systems of atoms. The fast Fourier transform (FFT) method is used for the spatial propagation of kinetic energy term. Equation (2) is integrated with the central difference method with the time step of 0.2×10^{-15} second. After integrating equation (2), the motions of ions are once stopped and equation (3) is propagated. The total calculation time is 400×10^{-15} second.

3. RESULTS AND DISCUSSIONS

Figure 1 shows a simple system of two atoms composed of two ions and two valence electrons for the analysis of interaction between light and matter. The light direction of light electric field is fixed to parallel to the connected line of the center of two atoms. In Table 1, system and light parameters, the dipole moment of ions q, potential parameters with delocalized electrons, light energy and light electric fields, are listed. The natural vibrational frequency of the atomic dimer without light irradiation corresponds to the light energy of 0.018 eV. The absorption cross-section spectra of the dimer has a peak at about 1.15 eV. The magnitude of light electric fields is chosen to be comparable to the value of a practical YAG laser. The light energy used in the calculation are 0.018, 0.03, 0.3, 1.15 and 2 eV and the light electric fields are 10^7, 10^8, 10^9 and 10^{10} V/m for each light energy. The dipole parameter q is chosen to be relatively larger than the usual permanent dipole moment of molecules (H–Cl: 3.7×10^{-20} C) so as to see the dipole effect in a short calculation time.

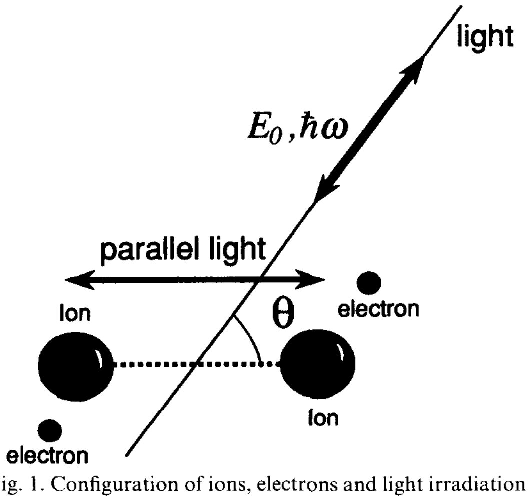

Fig. 1. Configuration of ions, electrons and light irradiation.

Table 1. Parameters of light and dimer system

Light		System	
Light energy $\hbar\omega$ [eV]	Light electric field E_0 [V/m]	Dipole moment q [C](q^*)	Excited potential V^{i-i}: (ε^*, σ^*)
0.018	10^7	0(0)	(1, 1)
0.03	10^8	3.7×10^{-19}(0.1)	(0.01, 1)
0.3	10^9	3.7×10^{-18}(1)	(0.5, 1.3)
1.15	10^{10}	—	—
2	—	—	—

Three cases are considered as the potential energy between ions with delocalized electrons; the one with the same ε and σ, with a smaller ε and the same σ and with a smaller ε and a larger σ than that of localized electrons. Under such various light and system conditions, the kinetic energy of the atomic dimer and valence electron states with light irradiation are calculated and discussed.

3.1. *Effect of light energy*

The effect of light energy on the dimer kinetic energy is discussed in this section. The dipole parameter of the system $q = 3.7 \times 10^{-18}$ C is used and parameters (ε, σ) of the ion potential with no irradiation are 1.625×10^{-19} J and 3.504×10^{-10} m, respectively. Ion potential parameters with delocalized valence electrons (ε, σ) are 8.125×10^{-20} J and 4.555×10^{-10} m. The dimer system with delocalized electrons has three parameters of $\varepsilon = 8.125 \times 10^{-20}$ J, $\sigma = 4.555 \times 10^{-10}$ m and $q = 3.7 \times 10^{-18}$ C of which non-dimensionalized values are $\varepsilon^* = 0.5$, $\sigma^* = 1.3$ and $q^* = 1$, respectively. In Fig. 2 cross marks show the light parameters used in this calculation. The horizonal axis of the figure is the light energy $\hbar\omega$ and the vertical axis is the light electric field E_0. The spectra of absorption cross section of this dimer calculated by equation (12) is also plotted in Fig. 2. The dotted line in Fig. 2 shows the light electric field which makes the potential energy change by more than 10% from that under no irradiation at 400 fs, the dot–dash line that of the kinetic energy change by more than 10% from that under no irradiation. The solid and dash lines are the light electric field which yields dissociation and ionization, respectively. When the electric field of the light is larger than these lines, it means that the corresponding phenomena can be observed at the final states of the atomic system. When the light electric field is more than 10^{10} V/m, the atomic dimer will be ionized and dissociated at all energy levels of light.

Contours of the electric charge distribution and ion positions under light irradiation are shown in Fig. 3 at 100, 200, 300 and 400 fs with the light energy of 0.018, 0.03, 0.3, 1.15 and 2 eV. The light electric field

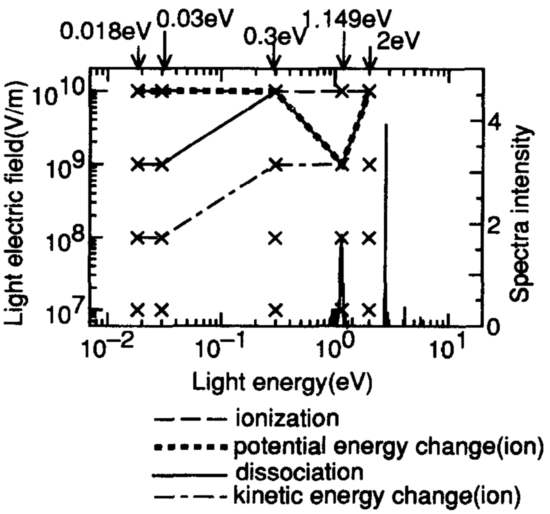

Fig. 2. Light parameters (light energy and light electric field), cross-section spectra of light absorption and final state of atomic dimer at 400 fs after light irradiation; the system parameters are ($\varepsilon^* = 0.5, \sigma^* = 1.3, q^* = 1$). The dash line, the dotted one, the solid one and the dot–dash one mean ionization, potential energy change (ion) from that in no irradiation case, dissociation and kinetic energy change (ion) from that in no irradiation case, respectively.

is 10^9 V/m in all cases. Dissociation of the atomic dimer into two atoms can be seen in the cases of light irradiation of 0.018, 0.03 and 1.15 eV.

Figure 4(a)–(c) shows time history of the ion kinetic energy, valence electron energy and ion distance under light irradiation, respectively, corresponding to the cases of Fig. 3. The kinetic energy of ions changes greatly in the case of 0.018, 0.03 and 1.15 eV but there is little change at 0.3 and 2 eV. Under light irradiation of 1.15 eV, the energy of valence electrons increases greatly because there is an absorption cross section peak at 1.15 eV. Ion distance increases gradually under light irradiation of 0.018 and 0.03 eV.

These results show that there are two mechanisms in dissociation and changes in the kinetic energy by light irradiation. The first one is of the infra-red light irradiation which acts mainly on dipole moment of the atomic dimer. The dipole moment fluctuates with light irradiation at the same frequencies as those of the atomic vibration to change the dimer vibrational motion and to increase the kinetic energy. This leads finally to dissociation of atomic dimer into two atoms. The characteristic time of dissociation or the increase in the kinetic energy is related to the light energy, light electric field, atomic initial states and dipole parameter q. The other mechanism is of visible light (at electron energy level) irradiation which excites valence electrons. Such electron excitation changes the potential between ions and leads to increase in the kinetic energy or dissociate the atomic cluster. In this process, it is rather important how the electron excitation changes the ion potential for kinetic energy change or dissociation.

3.2. *Effect of light electric field*

The calculation model and the system parameters are the same as those in Section 3.1. Charge distribution of valence electrons and ion positions are shown in Fig. 5 at 100, 200, 300 and 400 fs after light irradiation with the light electric field of 10^7, 10^8, 10^9 and 10^{10} V/m. The light energy is 0.018 eV for all cases. The atomic dimer is not dissociated within 400 fs at 10^7 and 10^8 V/m whereas it is dissociated at 10^9 V/m. It is also ionized and dissociated by 10^{10} V/m irradiation.

Figure 6(a)–(c) is the time history of the ion kinetic energy, valence electron energy and ion distance under light irradiation, respectively, at 0.018 eV light irradiation. At 10^8 V/m irradiation the kinetic energy begins to increase and the phase of atomic vibration to be changed from that under no irradiation. The valence electron energy fluctuates largely only in the case of 10^{10} V/m irradiation.

Figure 7(a)–(c) is the same as Fig. 6 with the light energy of 1.15 eV. Changes in the kinetic energy of ions and electron energy are seen when the light electric field is more than 10^8 V/m. From Figs 6–7, it is seen that stronger light electric fields can make the atomic dimer dissociate within shorter time of irradiation at all energy level of light. With the light electric field more than 10^{10} V/m, the atomic dimer is ionized and dissociated within 400 fs. The magnitude of light electric field dominates the characteristic time of both dissociation and increasing rate of the kinetic energy of ions.

3.3. *Effect of potential between ions*

In order to know the effect of ion potential with delocalized electrons on the evolution of the kinetic energy of ions, the motion of ions and electronic states are calculated with the potential parameters ($\varepsilon^*, \sigma^*, q^*$) of three cases, (1, 1, 1), (0.01, 1, 1) and (0.5, 1.3, 1). The results in the case of (0.5, 1.3, 1) are shown in Sections 3.1 and 3.2. In the cases of (1, 1, 1) and (0.01, 1, 1), the relationship between atomic state at 400 fs and light is shown in Fig. 8(a) and (b), respectively. The charge distribution of electrons and ion positions are plotted in Fig. 9. The atomic dimer is dissociated at 0.018 and 0.03 eV light irradiation. At 1.15 eV light irradiation, dissociation cannot be observed although the distribution of valence electrons is different from other cases. In the case of ($\varepsilon^*, \sigma^*, q^*$) = (0.5, 1.3, 1) in Fig. 2, the dissociation is observed at 1.15 eV light irradiation. This means that dissociation under light irradiation at the electron energy level is dominated by changes in the potential between ions with light absorption. These results show that the Lennard–Jones parameter σ is more important than ε for such dissociation and increase of the kinetic energy. Figure 10(a) and (b) show the time

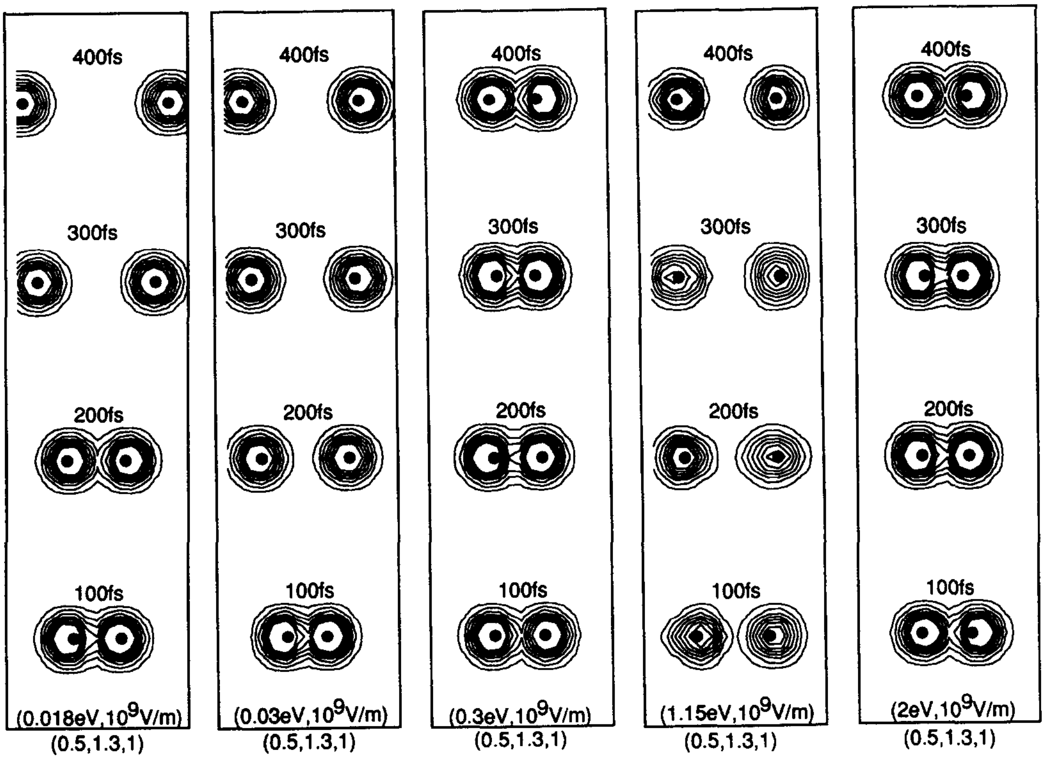

Fig. 3. Effect of light energy on electric charge distribution of electrons and ion position under light irradiation: the system parameters ($\varepsilon^* = 0.5, \sigma^* = 1.3, q^* = 1$); the light parameters ($\hbar\omega, E_0$) = (0.018 eV, 10^9 V/m), (0.03 ev, 10^9 V/m), (0.3 eV, 10^9 V/m), (1.15 eV, 10^9 V/m) and (2 eV, 10^9 V/m) from the left side, respectively.

history of the kinetic energy of ions in the case of $(\varepsilon^*, \sigma^*, q^*) = (0.01, 1, 1)$ and Fig. 11(a) and (b) in $(1, 1, 1)$. At 1.15 eV light irradiation, the kinetic energy changes in a different way from that at 2 eV irradiation, but any increase in the kinetic energy is not observed for both cases. The phase difference between two irradiations in Fig. 10 is larger than that in Fig. 11.

3.4. *Effect of dipole parameter q*

Effects of the dipole parameter q on the atomic motion are calculated by changing the value of the potential parameter as $(\varepsilon^*, \sigma^*, q^*) = (0.5, 1.3, 0)$, $(0.5, 1.3, 0.1)$. The results are shown in Fig. 12(a) for $(\varepsilon^*, \sigma^*, q^*) = (0.5, 1.3, 0)$ and Fig. 12(b) for $(0.5, 1.3, 0.1)$. The charge distribution and ion positions are plotted in Fig. 13. Dissociation is observed only at 1.15 eV light irradiation. In Fig. 14(a) and (b), the kinetic energy is shown for $(0.5, 1.3, 0)$ and in Fig. 15(a) and (b) for $(0.5, 1.3, 1)$. The ion kinetic energy evolves in a similar way in both cases. The kinetic energy hardly increases at 0.018 eV or 0.03 eV light irradiation. With the light irradiation (0.018 eV, 10^9 V/m), the kinetic energy tends to decrease. These results suggest that the dipole parameter q is very important for ion motion and dissociation under infra-red irradiation, but less appreciable under light irradiation at electron energy levels.

4. CONCLUSION

The quantum molecular dynamics method is developed to analyze the fundamental mechanism of light-to-heat absorption that the light is absorbed and converted to thermal energy in atomic systems of ion and electron. This method makes it possible for light irradiation (laser beam) to be dealt with as the electric field with two photon parameters, that is, the light angular frequency (ω) calculated from photon energy ($\hbar\omega$) and the light electric field (E_0) calculated from photon energy density or laser beam output. By using this method the relationships between the character of light irradiation and thermal effects on matters have been studied for a simple atomic system, two ions and two valence electrons.

Under infra-red light irradiation, increasing the magnitude of the light electric field is associated with increase in the kinetic energy and atomic dissociation which are mainly attributed to change in the atom dipole moment with light irradiation. The characteristic time of the kinetic energy increase is related to the light energy, light electric field and dipole moment q.

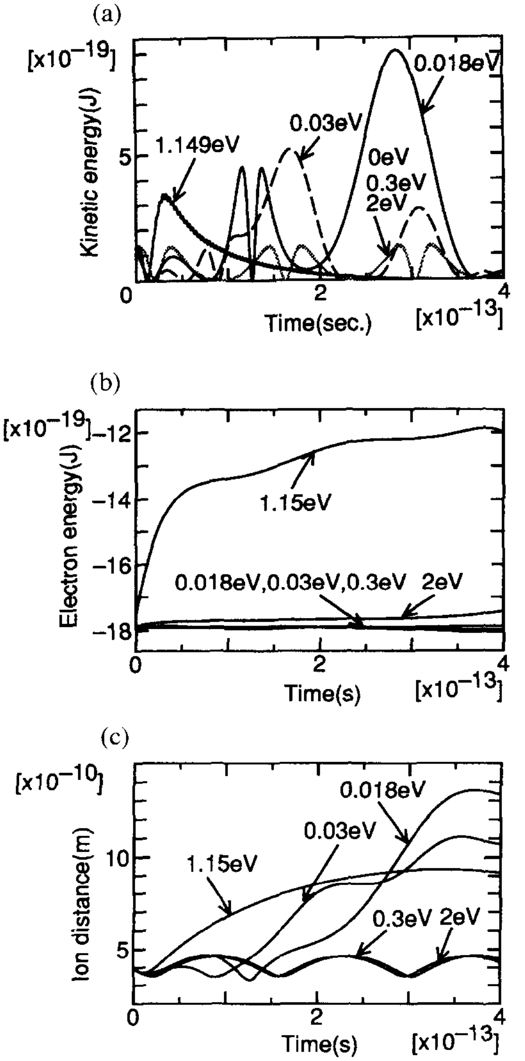

Fig. 4. Effect of the light energy on the time history of the kinetic energy of ions, electron energy and ion distance under light irradiation: the light parameters are the same as those in Fig. 3: (a) kinetic energy of ions; (b) energy of valence electrons; (c) distance between ions.

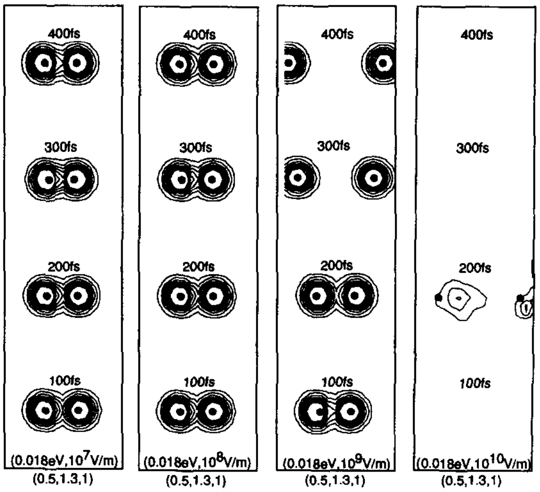

Fig. 5. Effect of light electric field on electric charge distribution of electrons and ion position under light irradiation: the system parameters ($\varepsilon^* = 0.5, \sigma^* = 1.3$, $q^* = 1$); the light parameters ($\hbar\omega, E_0$) = (0.018 eV, 10^7 V/m), (0.018 eV, 10^8 V/m), (0.018 eV, 10^9 V/m) and (0.018 eV, 10^{10} V/m) from the left side, respectively.

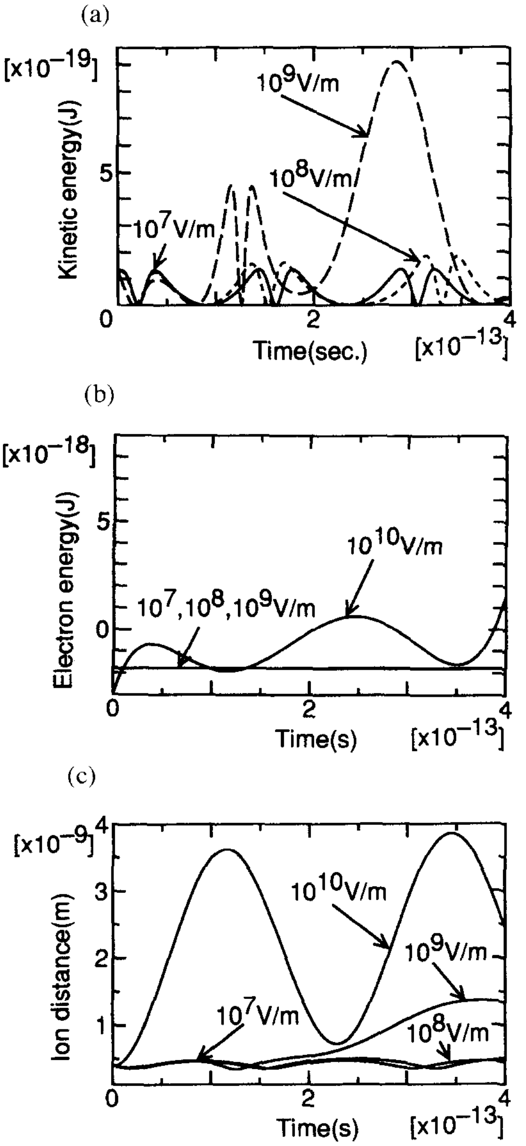
(a)
(b)
(c)

Fig. 6. Effect of light electric field on the time history of the kinetic energy of ions, electron energy and ion distance under light irradiation: the light parameters are the same as those in Fig. 5: (a) kinetic energy of ions; (b) energy of valence electrons; (c) distance between ions.

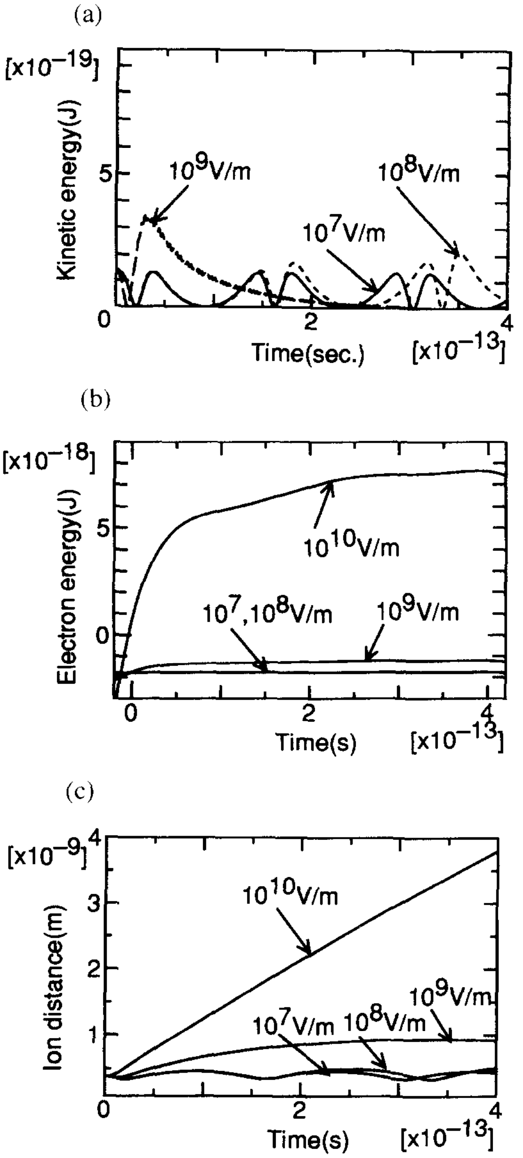
(a)
(b)
(c)

Fig. 7. Effect of light electric field on the time history of the kinetic energy of ions, electron energy and ion distance under light irradiation: the light parameters $(\hbar\omega, E_0) = (1.15$ eV, 10^7 V/m), (1.15 eV, 10^8 V/m), (1.15 eV, 10^9 V/m) and (1.15 eV, 10^{10} V/m): (a) kinetic energy; (b) energy of valence electrons; (c) distance between ions.

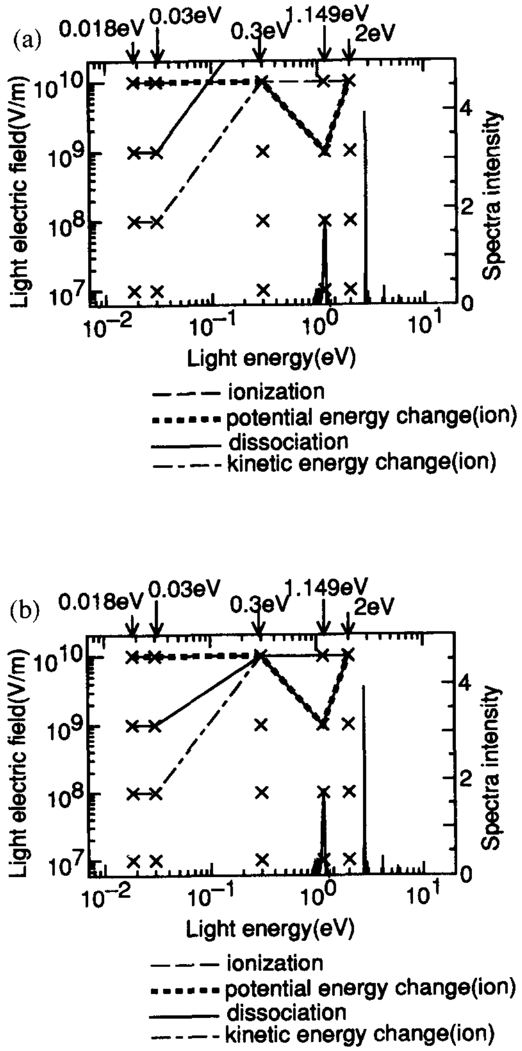

Fig. 8. Effect of ion potential on final state of atomic dimer at 400 fs after light irradiation: meaning of the lines are the same as in Fig. 2: (a) the system parameters ($\varepsilon^* = 1, \sigma^* = 1, q^* = 1$); (b) the system parameters ($\varepsilon^* = 0.01, \sigma^* = 1, q^* = 1$).

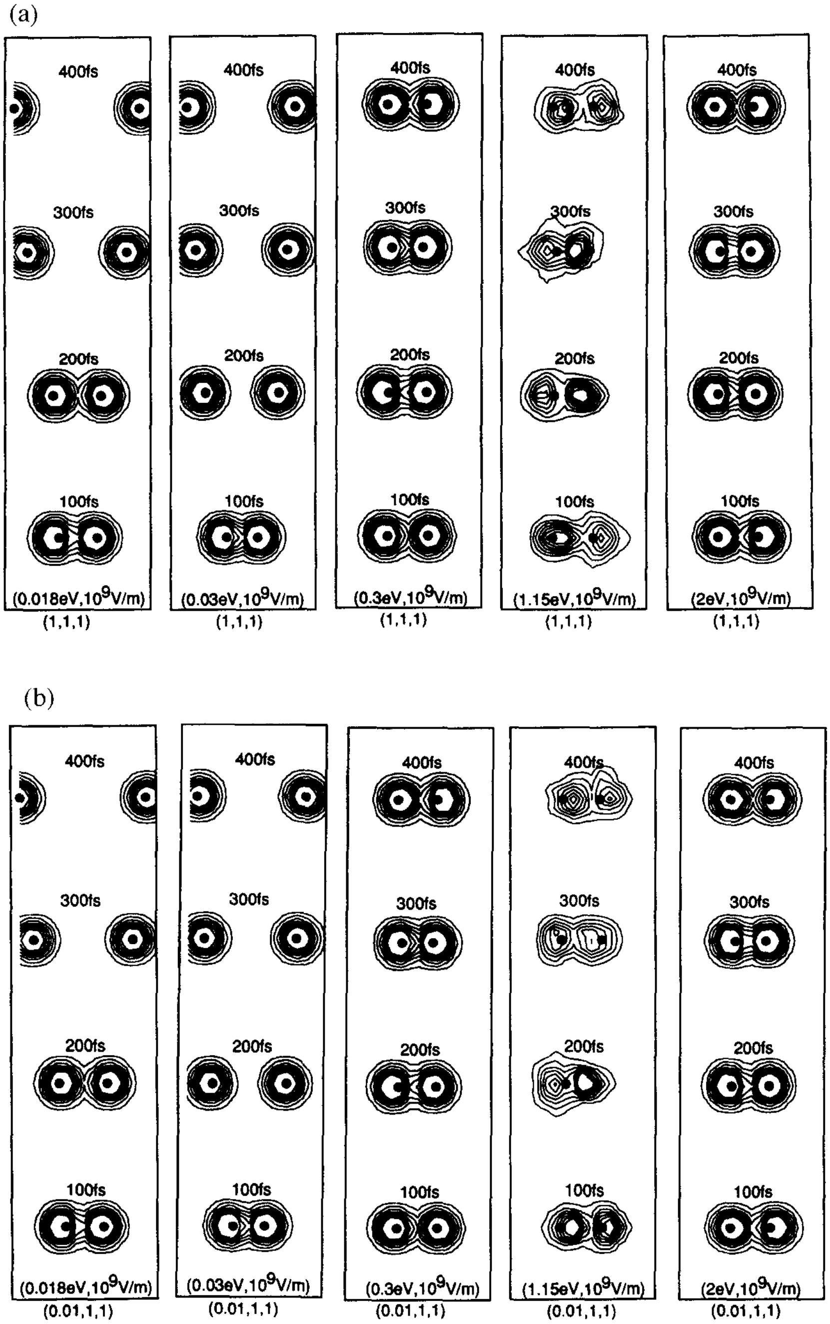

Fig. 9. Effect of ion potential on electric distribution of electrons and ion position under light irradiation: the light parameters ($\hbar\omega, E_0$) = (0.018 eV, 10^9 V/m), (0.03 eV, 10^9 V/m), (0.3 eV, 10^9 V/m), (1.15 eV, 10^9 V/m) and (2 eV, 10^9 V/m) from the left side, respectively: (a) the system parameters ($\varepsilon^* = 1, \sigma^* = 1, q^* = 1$); (b) the system parameters ($\varepsilon^* = 0.01, \sigma^* = 1, q^* = 1$).

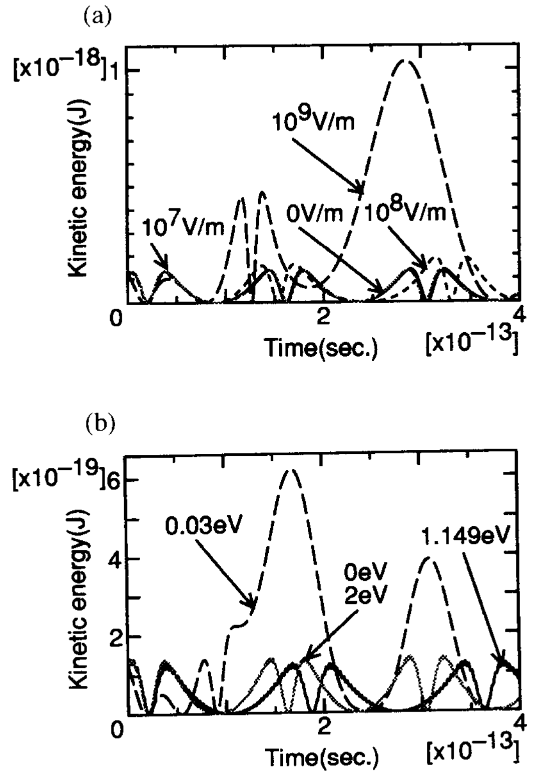

Fig. 10. Effect of ion potential on time history of the kinetic energy of ions under light irradiation : the system parameters ($\varepsilon^* = 0.01, \sigma^* = 1, q^* = 1$) : (a) the light parameters $(\hbar\omega, E_0) = (0$ V/m ; no irradiation), (0.018 eV, 10^7 V/m), (0.018 eV, 10^8 V/m) and (0.018 eV, 10^9 V/m) ; (b) the light parameters $(\hbar\omega, E_0) = (0$ eV ; no irradiation), (0.03 eV, 10^9 V/m), (1.15 eV, 10^9 V/m) and (2 eV, 10^9 V/m).

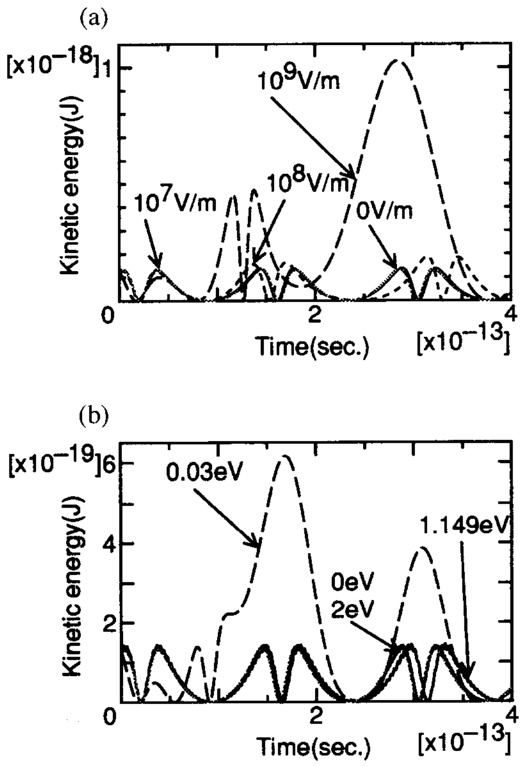

Fig. 11. Effect of ion potential on the time history of the kinetic energy of ions under light irradiation: the system parameters ($\varepsilon^* = 1, \sigma^* = 1, q^* = 1$) : (a) the light parameters $(\hbar\omega, E_0) = (0$ V/m ; no irradiation), (0.018 eV, 10^7 V/m), (0.018 eV, 10^8 V/m) and (0.018 eV, 10^9 V/m) ; (b) the light parameters $(\hbar\omega, E_0) = (0$ eV ; no irradiation), (0.03 eV, 10^9 V/m), (1.15 eV, 10^9 V/m) and (2 eV, 10^9 V/m).

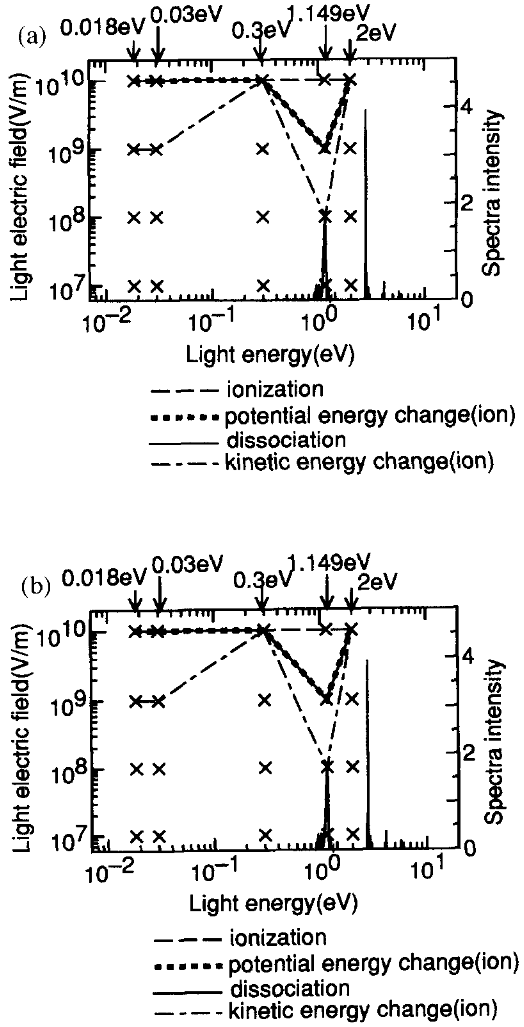

Fig. 12. Effect of dipole parameter q on final state of atomic dimer after light irradiation: meaning of the lines are the same as in Fig. 2: (a) the system parameters ($\varepsilon^* = 0.5, \sigma^* = 1.3, q^* = 0$); (b) the system parameters ($\varepsilon^* = 0.5, \sigma^* = 1.3, q^* = 0.1$).

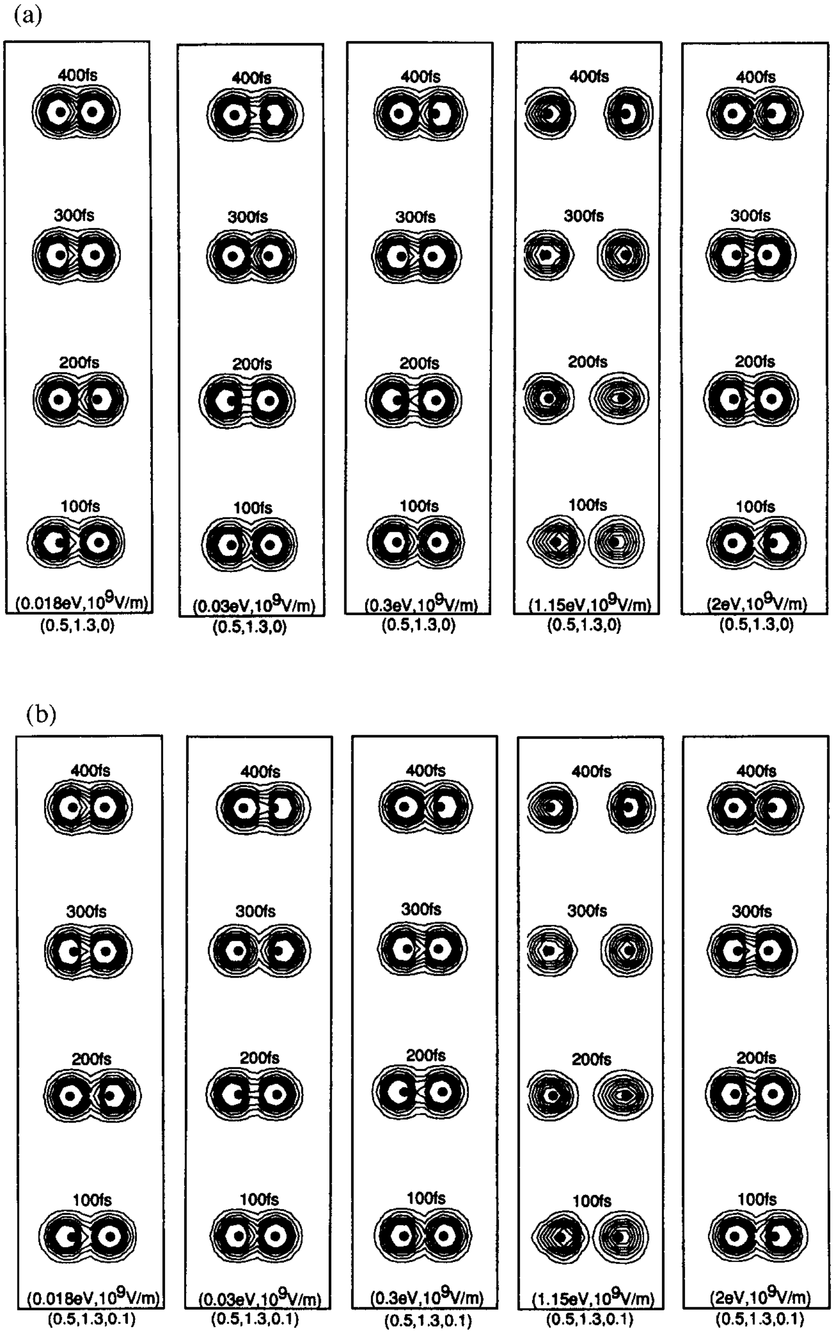

Fig. 13. Effect of dipole parameter q on electric charge distribution of electrons and ion position under light irradiation: the light parameters $(\hbar\omega, E_0) = (0.018\ \mathrm{eV}, 10^9\ \mathrm{V/m})$, $(0.03\ \mathrm{eV}, 10^9\ \mathrm{V/m})$, $(0.3\ \mathrm{eV}, 10^9\ \mathrm{V/m})$, $(1.15\ \mathrm{eV}, 10^9\ \mathrm{V/m})$ and $(2\ \mathrm{eV}, 10^9\ \mathrm{V/m})$ from the left side, respectively: (a) the system parameters $(\varepsilon^* = 0.5, \sigma^* = 1.3, q^* = 0)$; (b) the system parameters $(\varepsilon^* = 0.5, \sigma^* = 1.3, q^* = 0.1)$.

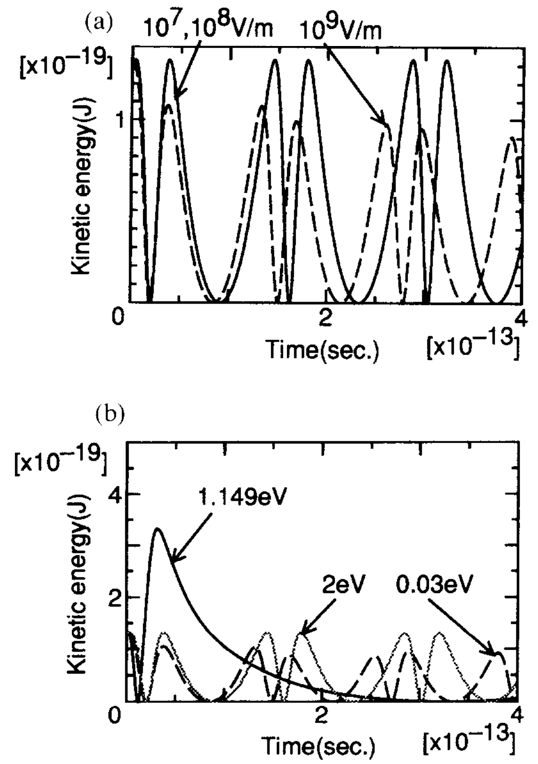

Fig. 14. Effect of dipole parameter q on the time history of the kinetic energy of ions under light irradiation: the system parameters ($\varepsilon^* = 0.5, \sigma^* = 1.3, q^* = 0$): (a) the light parameters $(\hbar\omega, E_0) = (0.018\ \text{eV}, 10^7\ \text{V/m})$, $(0.018\ \text{eV}, 10^8\ \text{V/m})$ and $(0.018\ \text{eV}, 10^9\ \text{V/m})$; (b) the light parameters $(\hbar\omega, E_0) = (0.03\ \text{eV}, 10^9\ \text{V/m})$, $(1.15\ \text{eV}, 10^9\ \text{V/m})$ and $(2\ \text{eV}, 10^9\ \text{V/m})$.

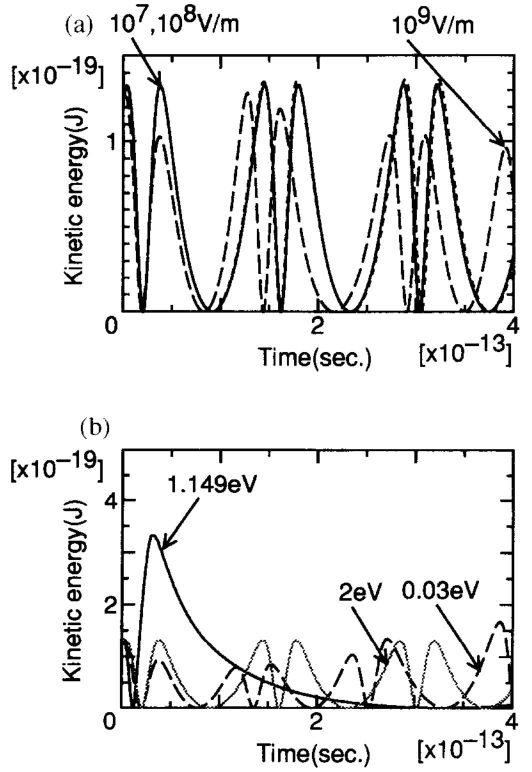

Fig. 15. Effect of dipole parameter q on the time history of the kinetic energy of ions under light irradiation: the system parameters ($\varepsilon^* = 0.5, \sigma^* = 1.3, q^* = 0.1$): (a) the light parameters $(\hbar\omega, E_0) = (0.018\ \text{eV}, 10^7\ \text{V/m})$, $(0.018\ \text{eV}, 10^8\ \text{V/m})$ and $(0.018\ \text{eV}, 10^9\ \text{V/m})$; (b) the light parameters $(\hbar\omega, E_0) = (0.03\ \text{eV}, 10^9\ \text{V/m})$, $(1.15\ \text{eV}, 10^9\ \text{V/m})$ and $(2\ \text{eV}, 10^9\ \text{V/m})$.

Under the irradiation of light at electron energy levels, increase in the kinetic energy and atomic dissociation occur mainly due to change in the ion potential parameter σ caused by electron excitation. They are not appreciably affected by the dipole moment q. For further understanding the fundamental mechanism of light absorption or radiative heat transfer, more strict calculations are needed under conditions taken into account the electron interactions as well as the ion excitation under light irradiation.

REFERENCES

1. Haug, K. and Metiu, K., *Journal of Chemical Physics*, 1991, **95**, 5670
2. Shibahara, M. and Kotake, S., *Proceedings of the ASME/JSME Thermal Engineering*, Vol. 1, pp. 461–466, Lahaina, Hawaii, 1995.
3. Engel, V., Schine, R., Hennig, S. and Metiu, H., *Journal of Chemical Physics*, 1990, **92**, 1.
4. Feit, M. D., Fleck, J. A. and Steiger, Q., *Journal of Computational Physics*, 1982, **47**, 412.