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Microscopic energy transport through photon-electron-phonon interactions during ultrashort laser ablation of wide bandgap materials Part Ⅰ: photon absorption

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Abstract The new phenomena induced by femtosecond lasers lead to the new area of ultrafast science. It is a significant challenge to explain the phenomena associated with complex non-equilibrium and non-linear processes. Although there is a growing body of experimental observation, a comprehensive model remains undeveloped. We review the challenges in understanding the photon absorption stage mainly for the femtosecond ablation of wide bandgap materials at the intensities of $10^{13}\sim10^{14}$ W/cm^2. Major opinions and challenges in ionization mechanisms are presented by primarily considering multiphoton ionization and avalanche ionization.

Key words femtosecond laser ablation; wide bandgap materials; photon-electron interactions; multiphoton ionization; avalanche ionization

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1 Introduction

Femtosecond lasers open wide-range and excit new possibilities in microfabrications of metals[1~4], polymers[5,6], semiconductors[7,8], ultrahard materials[9,10], transparent materials[11,12], and tissues[13] for automotive industry, pharmaceutical industry, process and automation technology, defense industry, aerospace industry, information technology, telecommunication technology, biotechnology, medicine industry, measurement and microscopy, environmental technology, etc[14~22].

Femtosecond pulse in some aspects fundamentally changes the laser-material interaction mechanism compared with a long pulse. A femtosecond laser can easily achieve very high peak power, which is powerful enough for full ionization of most solid material[23~25]. At such high intensities, seed free electrons are mainly generated by photoionization including multiphoton ionization and/or tunnel ionization[26], which is independent on the initial states of the target materials[27~29]. Hence, femtosecond laser ablation is much more deterministic and reproducible than long-pulse laser one[30,31]. Sub-diffraction structures can be achieved by choosing laser fluence slightly above the ablation threshold[32,33]. Femtosecond laser-material interaction is highly nonequilibrium[34~37]. Electrons are excited up to a few or tens of electron volts[38~40] corresponding to tens of femtoseconds[29], while the subsequent energy transferring from electrons to ions is of picosecond order[41,42]. Hence, the ultrafast pulse energy is mainly deposited in a small layer in the photon-electron interaction process in one pulse duration[43,44]. Heat conduction and hydrodynamic motion are significantly decreased during the laser irradiation. Thus, recast, thermal damage (microcracks), and heat-affected-zone (HAZ) are greatly reduced. The ablation depth of the femtosecond laser is typically on the order of 0.01～1 μm per pulse[45]. Hence, femtosecond laser ablation can be precise.

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The unique advantages of femtosecond lasers make them to be a very promising tool for the fabrication of wide bandgap materials that are difficult for conventional methods. Femtosecond ablation of wide bandgap materials is the main point of this paper. This is an active area with very significant scientific and engineering merits[11,29,46~49]. Bandgap is the energy difference between the top of the valence band and the bottom of the conduction band. There is no clear cutoff of bandgap to define the wide bandgap materials. In this paper, wide-band materials refer to materials with a bandgap greater than 3 eV, including some semiconductors, for example, ZnO (3.1 eV), SiC (3.0～3.25 eV), GaN (3.4 eV), silicon nitride (3.9～4.1 eV), and all dielectrics, such as, diamond (5.46～6.4 eV), AlN (6.2 eV), sodium chloride (7.5 eV), NaCl (8 eV), quartz (9～10 eV), silicon dioxide (9 eV), and sapphire (9.9 eV).

However, typical femtosecond laser ablation is low throughput with high photon cost, which is not suitable for typical industrial applications[50]. Further, the underlying physical mechanism of femtosecond laser ablation remains controversial and poorly understood[29,51~53]. Many approaches are employed to study femtosecond laser pulse ablation of wide bandgap materials. Most theoretical models are spatially averaged[29,54~58]. The ionization dynamics is coupled to an equation for the absorbed energy density derived by the Drude model. Multiple rate equations can also be found in the literature[59~61]. Another popular approach is based on laser propagation in the dielectric media by solving a single equation for either the laser electric field or vector potential in the retarded laser frame[62~65]. This approach can calculate the laser electromagnetic field in two dimensions, but it only considers the electromagnetic wave traveling in the forward direction and does not account for reflection from the plasma. Kinetic models based on the Boltzmann or Fokker-Planck equation such as those of Stuart *et al*. are more complex with better accuracy[47,51,66~68]. Resently Jiang *et al*. proposed a quantum multiscale model to understand ultrafast, non-equilibrium laser-material interactions from nm to mm and from fs to μs, as shown in Fig. 1[68]. The model is being used to reveal the fundamental science underlying ultrafast phase change mechanisms, which include three main parts: 1) understanding initial nonlinear laser radiation absorption through photon-electron and electron-electron interactions in a time scale from a few femtoseconds to picoseconds; 2) revealing the physics and chemistry of plasma generation and phase change through electron-ion interactions in a time scale from picoseconds to nanoseconds; and 3) investigating chemically-reactive fluid flow and radiation of expanding plasmas, and their interactions with environment and bulk materials in a few nanoseconds to hundreds of microseconds.

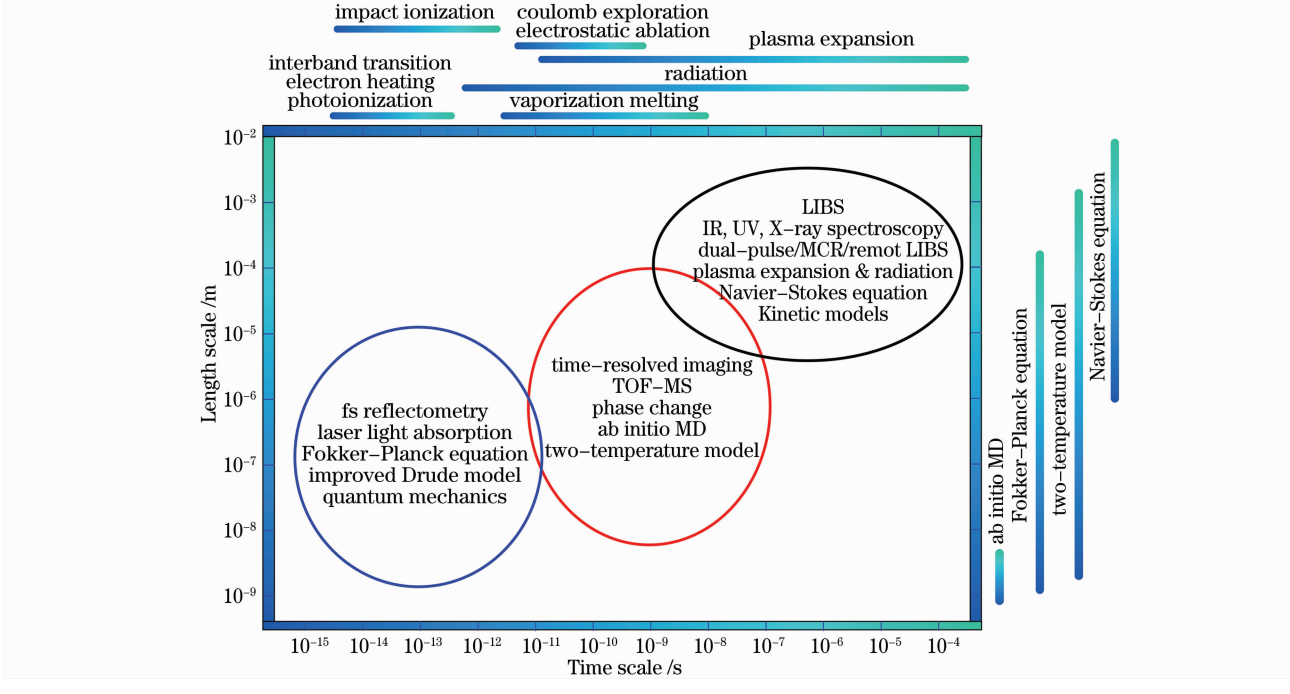

Fig. 1 Multiscale Modeling to understand the ultrafast, non-equilibrium laser-material interactions from nm to mm and from fs to μs

Recently there are some reviews on the femtosecond laser micro-/nano-fabrication and its applications with emphasis on the experiments[69~71], while this paper focuses on theoretical challenges in understanding energy transport during femtosecond laser ablation through photon-electron interactions mainly for wide bandgap materials at the intensity of $10^{13}\sim10^{14}$ W/cm^2. Avalanche ionization and multiphoton ionization are two major competing mechanisms that are considered for free electron generations in this section. After the critical density is created, the laser energy is absorbed mainly through inverse Brehmstrahlung and resonance absorption in a similar way as free electrons do in metals. Different views of the photon absorption process are reviewed in the next section.

2 Femtosecond laser pulse energy absorption

The energy transport in femtosecond laser ablation can be divided into two stages: 1) photon energy absorption by electrons, and 2) absorbed energy redistribution to lattice leading to material removals[48]. This stage separation is based on the assumption that photon-electron interaction completes in such a short time that the lattice temperature remains unchanged during the absorption of femtosecond pulse. This section reviews the absorption of laser energy through ionization and free electron heating.

2.1 Basic concepts in absorption of femtosecond laser energy

Linear photon absorption obeys the Beer-Lambert Law. Nonlinear absorption is significant at high electric fields and femtosecond pulse makes it easy to achieve high intensities. In femtosecond ablation, impact ionization (mainly avalanche ionization) and photoionization (multiphoton ionization and/or tunnel ionization) are major competing mechanisms for free electron generation[46,72].

As shown in Fig. 2, if the kinetic energy of a free electron becomes sufficiently high by absorbing photons, part of the energy may transfer to a bound electron by collisions to overcome the ionization potential[73] and produce two free electrons, which is called (collisional) impact ionization[47]. Consequently, the free electrons absorb photons and produce one more free electron from the bound electrons. Such a series of the impact ionization process is called avalanche ionization[46], where free electron density exponentially increases. Avalanche ionization strongly depends on free electron density and is sometimes assumed as linearly proportional to laser intensity[66]. Its efficiency is determined by competitions between energy gain through inverse Bremsstrahlung and energy loss through phonon emission.

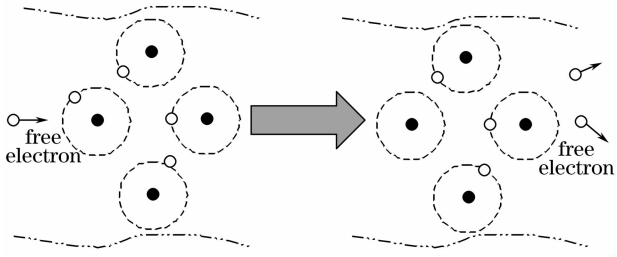

Fig. 2 Avalanche ionization consists of a series of impact ionizations

As shown in Fig. 3, in multiphoton ionization[74], several (N) photons with the energy of $h\nu$ at the wavelength of λ "simultaneously" strike a bound electron acting like a photon of $Nh\nu$ at the wavelength of λ/N due to very high photon flux (typically$>10^{31}$ cm$^{-2}\cdot$s^{-1}) of femtosecond lasers, i.e., a bound electron is freed from the valence band by absorbing several photons when the total energy of the absorbed photons is greater than the ionization potential[26,38]. This absorption process is actually achieved through metastable quantum state(s). N-photon ionization is an Nth order process, and thus the cross section is very small at low intensities ($<10^{12}$ W/cm^2). Hence, only if laser intensity (photon flux) is very high, multiphoton ionization can be significant. When the laser intensities are below 10^{12} W/cm^2, avalanche ionization is responsible for the ablation of wide bandgap materials and multiphoton ionization becomes significantly strong at higher intensities (typically $>10^{13}$ W/cm^2)[75~78]. However, when intensities are higher than 10^{15} W/cm^2, tunnel ionization becomes significant[29,51,66]. Actually, tunnel ionization or multiphoton ionization depends strongly on the value of the parameter γ, which is introduced by Keldysh[80]. The parameter is the ratio between the frequency of laser light ω and the frequency ω_t of electron tunneling through a potential barrier as following

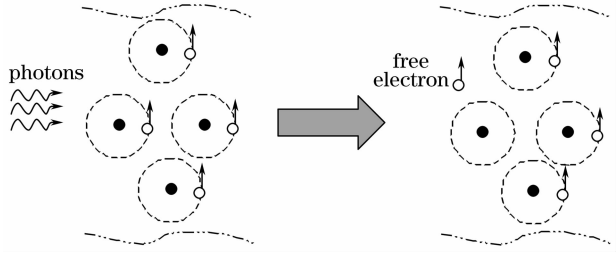

Fig. 3 Multiphoton ionization

$$\gamma = \frac{\omega}{\omega_t} = \frac{\omega\sqrt{2mI_p}}{e\varepsilon} = \frac{1}{2K_0 F}, \quad (1)$$

where $I_p = \kappa^2 me^4/\hbar^2$ is the ionization potential of the atomic level, ε is the amplitude of the electric wave field, $F = \varepsilon/\kappa^3\varepsilon_a$ is the reduced field, and $K_0 = I_p/\hbar\omega$ is the minimal number of photons required for ionization. Further, as a rule, atomic units $\hbar = m = e = 1$ are used, where m is the electron mass. Tunnel ionization takes place when $\gamma \ll 1$, while for $\gamma \gg 1$ the ionization is a multiphoton process. Tunnel ionization and multiphoton ionization belong to photoionization and sometimes they are called strong-electric-field ionizations[47,79,81]. Photoinization under femtosecond irradiation leads to metallic properties even for wide bandgap materials. Defects and impurities play a negligible role[58].

Laser ablation of a wide bandgap material is sometimes called laser-induced breakdown in which the material is firstly transformed into absorbing plasma with metallic properties and subsequent laser-plasma interaction causes the phase changes of bulk materials[27,30,38,48]. Buildup of free electrons is necessary to initialize the ablation. Free electron density is assumed to saturate at critical density, at which ablation occurs[66,76]. For femtosecond lasers, critical density is selected as the free electron density at which plasma oscillation frequency equals to the laser frequency. Jiang *et al*.[82] modified a plasma free electron model for metals and doped semiconductors to investigate highly ionized wide bandgap material because of the high density free electron generated by a femtosecond pulse. This model determines critical density by

$$n_{cr} = \frac{4\pi^2 c^2 m_e \varepsilon_0}{e^2\lambda^2}, \quad (2)$$

where m_e is the electron mass, c is the speed of light in vacuum, e is the electron charge, ε_0 is the electric permittivity of free space, and λ is the laser wavelength. n_{cr} is about 0.986×10^{21} cm^{-3} for 1064-nm wavelength. Similarly, Ref. [76] expresses the same equation as Eq. (2) in electrostatic unit (esu) as following

$$n_{cr} = \frac{\pi m_e c^2}{e^2\lambda^2}. \quad (3)$$

At critical density, transparent wide bandgap materials become totally opaque. A large percentage of absorbed energy is deposited in a very thin layer under strong intensity within a short time, which leads to the ablation of thin layer. The important role of critical density can be better understood by considering its impacts on optical properties of the ionized area. However, the selection of critical density using Eq. (2) is just a rough estimation. The relationship between the critical density and the pulsewidth should be clarified through analyzing the ionization mechanism, which remains a challenge.

2.2 Relative roles of avalanche ionization to multiphoton ionization

2.2.1 Multiphoton impact ionization

In femtosecond laser ablation of wide bandgap materials, the roles of multiphoton ionization and avalanche ionization in free electron generations are still controversial[27,29,48,51~53,78,83]. The early research on laser ablation of wide bandgap materials commonly assumed that avalanche ionization is responsible for the ablation[27,84~87]. Recently, some researchers believe that multiphoton ionization supplies seed electrons while avalanche ionization is still responsible for femtosecond ablation[32,39,46,66,72,83,88,89]. Perry *et al*. named this process multiphoton impact ionization[77]. This opinion is in accord with the experiments of Du *et al*.[88] on the ablation of fused silica using 150-fs～7-ns, 780-nm laser with the simplified analysis using the equation of Bloembergen[84].

Another evidence of multiphoton impact ionization is the experiments of Pronko *et al*. on the ablation of silicon using 80-fs～9-ns, 786-nm～1.06-μm lasers[46]. Their experiments show that the threshold of 1064 nm is always below that of 786 nm. Pronko *et al*. concluded that avalanche ionization still dominated the ablation[46], because in near IR range, longer wavelength radiation was more effective in producing avalanche ionization[90]. However, the wavelength dependence of the femtosecond ablation threshold is controversial[11,32,52,72,84,90].

Recently, Jia *et al*. studied the time-resolved electronic excitation using a pump and probe system and found that the reflectivity increased rapidly in the latter half of the pump pulse duration, which supported that impact ionization played an important role in the generation of conduction band electrons[11]. Xu *et al*. obtained the similar result that avalanche ionization played the dominant role in the femtosecond laser-induced breakdown in MgO near the damage threshold[91]. Sun *et al*. in the same group concluded that both multiphoton ionization and avalanche ionization played important roles in the femtosecond laser-induced damage in MgF_2[92]. Rethfeld *et al*. also studied the role of impact ionization as compared to multiphoton ionization by modeling and concluded that the contribution of impact ionization depended on the product of intensity

and duration of the laser pulse, thus on the laser fluence[60]. Petrov *et al*. believe that some researchers overestimates their photoionization rate by a factor of $\sim 20 \sim 100$ for intensities between 1×10^{13} W/cm^2 and 1×10^{14} W/cm^2, which favors photoionization as the dominant mechanism[93]. They concluded that the dominant process for electron multiplication depended only on the laser fluence F. For $F<0.4$ J/cm^2, the cumulative optical field ionization rate exceeds the impact ionization rate, while for $F>0.4$ J/cm^2, the impact ionization is the dominant ionization mechanism. Since for laser pulse duration exceeding 20 fs, the threshold for dielectric damage of SiO_2 is at least $F_{thr}>1.5$ J/cm^2[29], the primary cause for typical dielectric damage is an electron avalanche due to the impact ionization.

Perry, Stuart, and their colleagues developed theories for multiphoton impact ionization based on the kinetic equation and experiments on the ablation of dielectrics at 1053 nm, 852 nm, and 526 nm at the pulsewidth of 100 fs$\sim$1 ns[51,66,76]. In their theory, the free electron density distribution, $n_e(\varepsilon,t)$, is described by the Fokker-Plank equation as following[51]

$$\frac{\partial}{\partial\varepsilon}\Big[R_J(\varepsilon,t)n_e(\varepsilon,t)-\gamma(\varepsilon)E_p n_e(\varepsilon,t)- D(\varepsilon,t)\frac{\partial n_e(\varepsilon,t)}{\partial\varepsilon}\Big]+\frac{\partial n_e(\varepsilon,t)}{\partial t}=S(\varepsilon,t), \quad (4)$$

where ε is the electron kinetic energy, t is time, $R_J(\varepsilon,t)$ is the heating rate of electrons, $\gamma(\varepsilon)$ is the rate of electron-phonon energy transfer to the lattice, E_p is the energy of the typical phonon, $D(\varepsilon,t)$ is the diffusion coefficient, and $S(\varepsilon,t)$ is the source and sinks of electrons. The terms within the square bracket of Eq. (4) represent the electron distribution change because of Joule heating $R_J(\varepsilon,t)n_e(\varepsilon,t)$, the inelastic scattering of phonon $\gamma(\varepsilon)E_p n_e(\varepsilon,t)$, and electron energy diffusion $D(\varepsilon,t)\dfrac{\partial n_e(\varepsilon,t)}{\partial\varepsilon}$. The heating rate of electrons is taken as

$$R_J(\varepsilon,t)=\frac{\delta(\varepsilon)}{3}E^2(t), \quad (5)$$

where $E(t)$ is the electric field, and $\delta(\varepsilon)$ is the ac conductivity of an electron described by

$$\delta(\varepsilon)=\frac{e^2\tau_m(\varepsilon)}{m^*[1+\omega^2\tau_m^2(\varepsilon)]}, \quad (6)$$

where $1/\tau_m$ is the energy-dependent and electron-phonon transport scattering rate, m^* is the effective mass of an electron that is about the rest mass of an electron, and ω is the laser frequency. The diffusion coefficient is given by

$$D(\varepsilon,t)=2\varepsilon R_J(\varepsilon,t). \quad (7)$$

The term on the right of Eq. (4), $S(\varepsilon,t)$, the source and sinks of electrons, includes the impact ionization term $S_{imp}(\varepsilon,t)$ and photoionization term $S_{pi}(\varepsilon,t)$.

$$S(\varepsilon,t)=S_{imp}(\varepsilon,t)+S_{pi}(\varepsilon,t)= 4n_e(2\varepsilon+U_{IP},t)\nu_I(2\varepsilon+U_{IP})- n_e(\varepsilon,t)\nu_I(\varepsilon)+S_{pi}(\varepsilon,t), \quad (8)$$

where U_{IP} is the ionization potential, and $\nu_I(\varepsilon)$ is the impact ionization rate described by the Keldysh impact formula as following[80]

$$\nu_I(\varepsilon)=\chi\left(\frac{\varepsilon}{U_{PI}}-1\right)^2, \quad (9)$$

with χ being a proportionality constant. The source term of impact ionization[51,72,66,76,77], and that of photoionization[72,79,94] in Eq. (8), have been extensively studied.

If energy deposited into electrons is much more than that to lattice during the pulse, a widely accepted simplified model is derived from Eqs. (4)$\sim$(9) and described by[51]

$$\frac{dn_e(t)}{dt}=\beta(I)n_e(t)+P(I), \quad (10)$$

where $\beta(I)$ is the impact ionization term, and $P(I)$ is the photoionization term. The loss term because of electron diffusion and recombination is neglected in Eq. (10).

One of the major issues of this model is how to estimate impact ionization rate and photoionization rate. Stuart *et al*.[66] assumed that 1) as soon as the kinetic energy of an electron reached the critical energy, it produced another electron by impact ionization and both electrons became zero kinetic energy, which was so-called "flux-doubling" condition, and 2) the shape of electron distribution remained unchanged during the avalanche ionization. Under the two assumptions, at high laser intensities, avalanche rate can be assumed linearly proportional to laser intensity as following

$$\beta=\alpha_i I(t), \quad (11)$$

where α_i is a constant. In the case that the bandgap of the material is not too much greater than the photon energy and there is no intermediate resonance, the photoionization rate can be expressed as[66]

$$P(I)=\delta_N\left(\frac{I}{\hbar\omega}\right)^N N_s, \quad (12)$$

where δ_N is the cross section of N-photon ionization with a unit of cm^{2N}s^{N-1} that depends on the location of all other electrical states of the system, and N_s is the solid atom density. Equation (12) does not consider tunnel ionization.

The result of Eq. (10) has been proven effectively by comparing its results with the numerical solution of Eq. (4)[66]. Figure 4 shows that the ava-

lanche ionization is mainly responsible for the ablation, while multiphoton ionization just supplies seed free electrons in the ablation of fused silica by a 100-fs, 1053-nm pulse at 10^{13} W/cm^2[76]. Note that the axis of electron density is logarithmic.

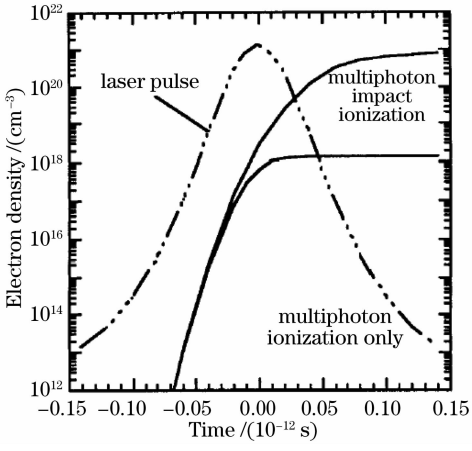

Fig. 4 Calculated free electron density for the ablation of fused silica using a 100-fs, 1053-nm pulse at 10^{13} W/cm^2[76]

As shown in Fig. 5, the threshold predictions based on the flux-doubling model are in agreements with the experiments conducted by the same group[76]. The methodology used to estimate the threshold is detailed in section 2.4. Although Perry's group assumed that multiphoton impacted ionization in their calculations[51,66,76,77], and the results demonstrated that at a pulsewidth shorter than 100-fs, the predicted threshold approached the prediction by multiphoton ionization alone represented by the dashed lines in Fig. 5. In this sense, the free electron generation dominated by multiphoton ionization can be considered as a specific case of the ablation by multiphoton impact ionization, where multiphoton ionization is so strong that the critical density is created before that avalanche ionization significantly initializes.

However, the model of Stuart *et al*.[51] fails to

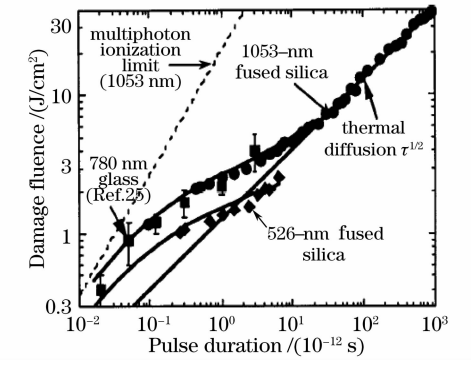

Fig. 5 Ablation threshold of fused silica at different pulse widths[76]

explain the increase in threshold fluence with the decrease of pulse width (< 1 ps)[72]. Also, the model fails to interpret the incubations effects on ablation threshold of dielectrics as a function of the delay between dual beams[83,95]. Li *et al*. added a decay term into the model to fit their experiment results as following[83]

$$\frac{\mathrm{d}n_e}{\mathrm{d}t} = \alpha_i I(t) n_e + \delta_N \left(\frac{I}{\hbar\omega}\right)^N N_s - \frac{n_e}{\tau}, \quad (13)$$

where τ is the decay time constant. However, this improvement was challenged by Petite *et al*.[95]. They explained the fast decay was a consequence of the self-trapping, which disagreed with the assumption that seed electrons left by the first pulse contributed the avalanche ionization in the second beam.

2.2.2 Thornber's model for avalanche rate and Keldysh's model for photoionization rate

As mentioned before, simplified methods of Eqs. (11) and (12) for avalanche rate and photoionization rate are valid for limited conditions. Thornber's model and Keldysh's theory can calculate the avalanche rate and photoionization rate, respectively, which is valid for many cases[72]. Thornber's model does not need the assumption of the linear proportionality between laser intensity and avalanche rate described by[72,96]

$$\beta(E) = \frac{v_s eE}{E_g} \exp\left[-\frac{E_I}{E(1+E/E_{phonon})+E_{kT}}\right], \quad (14)$$

where v_s is the saturation drift velocity ($\sim 2\times 10^7$ cm/s), E_g is the bandgap energy, and E_I, E_{phonon}, and $E_{kT} = E_I kT/E_g$ are the fields for carriers to overcome the decelerating effects of ionization scattering, optical phonon scattering, and thermal scattering in one mean free path, respectively.

Keldysh's theory[80] can be used to calculate the photoionization rate as a function of the laser electric field with the consideration of tunnel ionization as following[72]

$$P(E) = \frac{2\omega}{9\pi}\left(\frac{\omega m}{\sqrt{\gamma_1}\hbar}\right)^{3/2} Q(\gamma, x) \times \exp\left\{-\pi\langle x+1\rangle \frac{K_1(\gamma_1)-E_2(\gamma_1)}{E_2(\gamma_2)}\right\}, \quad (15)$$

where ω is laser frequency, and m is the reduced mass described by

$$m = \frac{m_e m_h}{m_e + m_h}, \quad (16)$$

where m_e is the mass of electron and m_h is the mass of the hole. In Eq. (15), γ is Keldysh's parameter in a solid,

$$\gamma = \frac{\omega\sqrt{mE_g}}{eE}, \tag{17}$$

and,

$$\gamma_1 = \frac{\gamma^2}{1+\gamma^2}, \quad \gamma_2 = 1-\gamma = \frac{1}{1+\gamma^2}, \tag{18}$$

$$x = \frac{2}{\pi}\frac{E_g}{\hbar\omega}\frac{\sqrt{1+\gamma^2}}{\gamma}E_2\left(\frac{1}{1+\gamma^2}\right), \tag{19}$$

$$Q(\gamma,x) = \sqrt{\frac{\pi}{2K_1(\gamma_2)}}\times \sum_{n=0}^{\infty}\exp\left\{-n\pi\frac{K_1(\gamma_2)-E_2(\gamma_2)}{E(\gamma_1)}\right\}\times \Phi\left\{\frac{\pi}{2}\sqrt{\frac{(2\langle x+1\rangle-2x+n)}{K_1(\gamma_2)E_2(\gamma_2)}}\right\}, \tag{20}$$

where K_1, E_2 are the complete elliptic integral of the first kind, and second kind respectively, $\langle z\rangle$ is the integer part of the number of z and

$$\Phi(z) = \int_0^z \exp(y^2-z^2)\,\mathrm{d}y. \tag{21}$$

However, the prediction of Keldysh's model for multiphoton ionization is substantially higher than the measurements by Lenzner *et al.*[29] on the ablation of dielectrics using 5-ps ~ 5-fs and 780-nm lasers.

2.2.3 Probability form for ionization rates

To make the relative friction of avalanche ionization to multiphoton ionization clearer, Gamaly *et al.*[52] express the free electron density as

$$\frac{\mathrm{d}n_e}{\mathrm{d}t} = \beta n_e + p_{mpi}N_s, \tag{22}$$

where N_s is the solid atom density, β is the time dependent probability of impact ionization, and $p_{mpi} = P/N_s$, is the time dependent of probability of multiphoton ionization[26,97,98]. Gamaly *et al.*[52] estimate β and p_{mpi} by

$$\beta = \frac{\varepsilon_{osc}}{U_{PI}}\left(\frac{2\omega^2\nu_{eff}}{\omega^2+\nu_{eff}^2}\right), \tag{23}$$

$$p_{mpi} = \omega N^{3/2}\left(\frac{\varepsilon_{osc}}{2U_{PI}}\right)^N, \tag{24}$$

where ε_{osc} is the electron quiver (oscillation) energy in the laser field, N is the minimum number of photons needed to overcome the bandgap in multiphoton ionization, and ν_{eff} is the effective collision frequency. Eqs.(23) and (24) show that the relative role of avalanche ionization to multiphoton ionization significantly depends on the electron quiver energy and ionization potential. If $\varepsilon_{osc} > U_{PI}$, multiphoton ionization dominates the process. Oscillation energy, ε_{osc}, in the unit of eV, is expressed in a scaling form as[52]

$$\varepsilon_{osc} = 9.3(1+\alpha_{pol}^2)\frac{I}{10^{14}}\lambda^2, \tag{25}$$

where α_{pol} is the coefficient for the beam polarization (for the circular, $\alpha_{pol}=1$, and for the linear polarization, $\alpha_{pol}=0$), I is in the unit of W/cm^2, and λ is in the unit of μm.

If laser intensity is greater than 10^{14} W/cm^2, it is obvious that $\varepsilon_{osc} > U_{PI}$ and multiphoton ionization dominates ablation process for most materials. For instance, silicon ablated by 1064-nm and 100-fs pulse at 2×10^{13} W/cm^2, avalanche ionization is responsible for the ablation, while at 10^{14} W/cm^2, multiphoton ionization dominates the process[52]. The predication of Gamaly *et al.*[52] is supported by the recent experiments on imaging femtosecond laser-induced electronic excitation in glass by Mao *et al.*[78]。They measured the evolution of laser-induced electronic plasma by a femtosecond time-resolved imaging technique at the intensity range of $5\times10^{12}\sim10^{14}$ W/cm^2.

2.2.4 Ablation dominated by multiphoton ionization

Some researchers[47,48,99] discovered that multiphoton ionization actually dominated absorption at the laser intensities on the order of 10^{14} W/cm^2. Using a frequency-domain interferometry to measure the relative phase of the reference and probe beams, Quéré *et al.* discovered no sign of avalanche ionization in the ablation of SiO_2 (9.0 eV), Al_2O_3 (8.8 eV), and MgO (7.65 eV), by a 790-nm, 60-fs laser[99]. They concluded that multiphoton ionization dominated in femtosecond laser ablation of dielectrics. Using the time dependent Boltzman equation, Kaiser *et al.*[47] demonstrated that at a laser electric field of 150 MV/cm, avalanche ionization was of minor importance for laser pulsewidths below 100 fs and only at pulsewidths above 200 fs avalanche ionization became as important as multiphoton ionization, as shown in Fig.6.

2.3 Electron heating

At intensities above 10^{14} W/cm^2, it takes just a few femtoseconds to reach the critical electron density by ionizations under femtosecond pulse[52]. After the critical density is created, the laser energy is absorbed mainly through inverse Brehmstrahlung and resonance absorption mechanism[53,100,101,102] in a similar way as free electrons do in metals. The refractive index can be calculated by[52]

$$r = \sqrt{\frac{\omega_{pe}}{2\omega}\left(1+\frac{\omega^2}{\omega_{pe}^2}\right)^{-1}}, \tag{26}$$

where ω_{pe} is electron plasma frequency. The absorption coefficient can be estimated by Fresnel formula as following[103]

$$\alpha = \frac{4r}{(1+r)^2+r^2}. \tag{27}$$

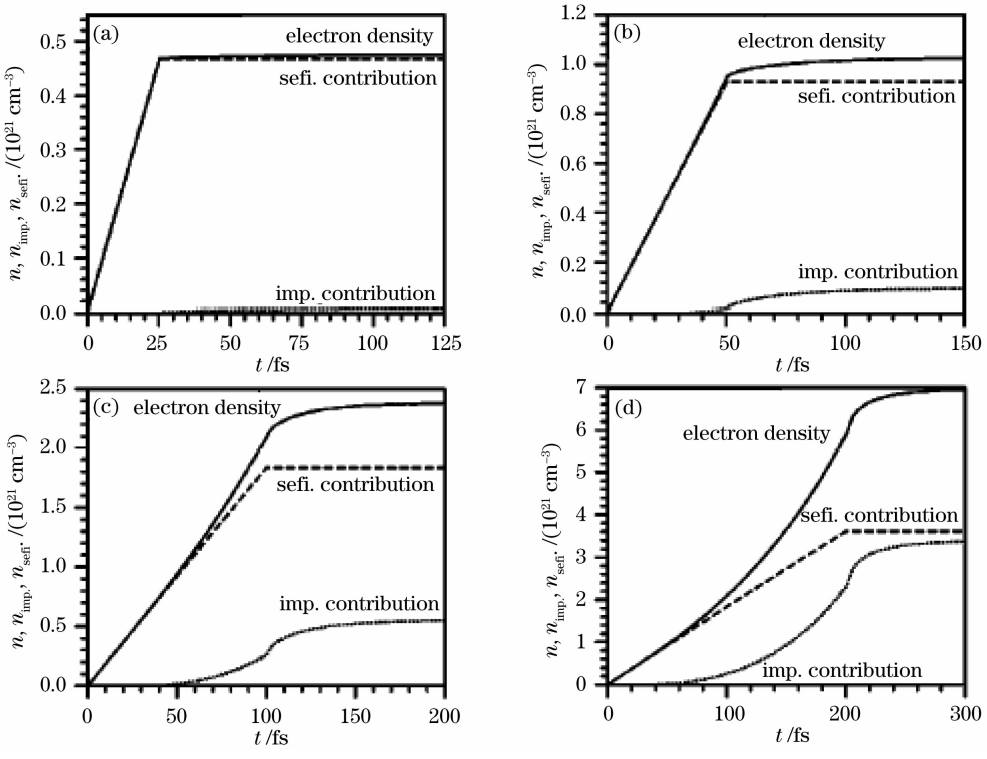

Fig. 6 Time dependent of free electron contribution by avalanche ionization (impact ionization presented as imp.) and photoionization (strong-electric-field ionization presented as sefi.) at 150 MV/cm with different pulsewidths. (a) 25 fs; (b) 50 fs; (c) 100 fs; (d) 200 fs[47]

During femtosecond pulse duration, electrons have no time to transfer energy to ions or out of the bulk material. Most of the pulse energy is firstly deposited into electrons in a small depth by the laser electric field. The laser electric field can be found as a solution to the Maxell equation coupled to the material equation. The solution is straightforward when the material parameters are constants in time and space and independent on the incident intensity. In this case, the interaction falls into the framework of well-known skin effects. The skin layer can be estimated by

$$l_s \approx \alpha \frac{c}{2\omega}\left(1+\frac{1}{r}+\frac{1}{2r^2}\right). \tag{28}$$

Electron conduction time t_h can be calculated by[104]

$$t_h = \frac{l_s^2}{\kappa} = \frac{3l_s^2}{l_e v_e}, \tag{29}$$

where κ is the coefficient of thermal diffusion, and l_e and v_e are electron free mean paths and velocity, respectively. The energy conservation of the electron energy in the process is described as[105]

$$c_e(T_e)n_e\frac{\partial T_e}{\partial t} = -\frac{\partial Q}{\partial z}, \tag{30}$$

where c_e is the specific heat of free electrons, and Q is the absorbed energy flux in the skin layer described by

$$Q = \alpha I \exp\left(-\frac{2z}{l_s}\right), \tag{31}$$

with α being the absorption coefficient. The solution to the Eq.(30) is simple under the assumptions of that free electron density n_e, absorption coefficient α, and skin layer l_s, are all time independent. However, all the three quantities are actually the functions of the laser intensity and time, which reduces the effectiveness of the skin-layer model. Furthermore, free electrons are not in equilibrium states, which has been demonstrated by experiments[106,107]. Rethfeld *et al.*[25] investigated the non-equilibrium dynamics of the electrons in femtosecond laser ablation of metals using a full Boltzmann collision integral free of phenomenological parameters. They found that the laser energy absorption was well described by a plasma-like absorption term after the critical density was created. In a quantum plasma model, Jiang *et al.*[68] considered the time and space dependent optical properties of highly ionized area in wide bandgap materials that turned out to be very critical for the ablation.

3 Conclusions

The new phenomena induced by femtosecond lasers lead to the new area of ultrafast science. It is a significant challenge to explain the phenomena as-

sociated with complex non-equilibrium and non-linear processes. This paper reviews recent developments of theoretical challenges in energy absorption stage during the femtosecond ablation of wide bandgap materials at the intensities of $10^{13}\sim 10^{14}$ W/cm^2. After the critical density is created, the laser energy is absorbed mainly through inverse Brehmstrahlung and the resonance absorption mechanism. Free electron density, reflectivity, absorption coefficient, and skin layer length are all time, space, and intensity dependent. These facts make the skin-effect model inapplicable to describe the electron heating process. An integrated multiscale physico-chemical modeling should be established to understand the ultrafast, non-equilibrium laser-dielectrics interactions from nanometer to milimeter and from femtosecond to microsecond.

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