Enhanced energy deposition and carrier generation in silicon induced by two-color intense femtosecond laser pulses

Mizuki Tani,1,† Kakeru Sasaki,1 Yasushi Shinohara,1,2 and Kenichi L. Ishikawa1,2,3,‡

1Department of Nuclear Engineering and Management, Graduate School of Engineering, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan
2Photon Science Center, Graduate School of Engineering, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan
3Research Institute for Photon Science and Laser Technology, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-0033, Japan

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We theoretically investigate the optical energy absorption of crystalline silicon subject to dual-color femtosecond laser pulses, using the time-dependent density functional theory (TDDFT). We employ the modified Becke-Johnson (mBJ) exchange-correlation potential which reproduces the experimental direct bandgap energy $E_g$. We consider situations where the one color is in the ultraviolet (UV) range above $E_g$ and the other in the infrared (IR) range below it. The energy deposition is examined as a function of mixing ratio $\eta$ of the two colors with the total pulse energy conserved. Energy transfer from the laser pulse to the electronic system in silicon is dramatically enhanced by simultaneous dual-color irradiation and maximized at $\eta \sim 0.5$. Increased is the number of generated carriers, not the absorbed energy per carrier. The effect is more efficient for lower IR photon energy, or equivalently, larger vector-potential amplitude. As the underlying mechanism is identified the interplay between intraband electron motion in the valence band (before excitation) driven by the IR component and resonant valence-to-conduction interband excitation (carrier injection) induced by the UV component. The former increases excitable electrons which pass through the $k$ points of resonant transitions. The effect of different multiphoton absorption paths or intraband motion of carriers generated in the conduction band play a minor role.

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I. INTRODUCTION

The ultrashort laser ablation of semiconductor and dielectric materials [1–4] has been attracting increasing attention due to both broad scientific interest and industrial application [5, 6]. Its advantages include high efficiency and quality [2, 7] thanks to the suppression of heat-affected-zone. The laser ablation is initiated by the absorption of optical energy to electrons. Then, irreversible damage is left on the material surface when the energy is subsequently transferred to the lattice [4], the carrier density reaches the critical density where the plasma frequency is identical to the laser frequency [8], or the interatomic forces are strongly modified owing to massive carrier creation [9–11]. Thus, the fundamental understanding of energy transfer from laser pulses to electrons is critical to further improve the efficiency of laser micro-machining [12–13].

There have been studies reporting that the use of synthesized dual-color laser pulse pairs enables highly efficient laser ablation of transparent materials, compared to single-color irradiation [14–23]. In Ref. [14, 15], the high efficiency observed under dual-color nanosecond ultraviolet (UV) lasers is attributed to the excited-state absorption (ESA) mechanism where the shorter wavelength component breaks the Si-O covalent bond of fused silica, which increases the absorption efficiency of the longer wavelength component. References [16, 17] have reported that the shorter wavelength laser excites valence electrons into defect or impurity energy levels and, then, that the longer wavelength laser promotes them from the localized levels to conduction states, even if the former alone cannot directly excite valence electrons into the conduction band. References [18, 19] have shown that the combination of picosecond or nanosecond infrared (IR) laser and its 2nd/3rd harmonics also improves ablation efficiency of silicon and 3C-SiC and attributed it to a mechanism where the UV laser excites valence electrons into the conduction band, which are then heated by the IR pulse. References [20, 21] have shown by experiments and analyses based on rate equations that a pair of femtosecond IR laser and its third harmonic UV can reduce ablation threshold of fused silica thanks to a defect state that can be reached by a single UV photon absorption. It has also been reported that the simultaneous use of femtosecond IR laser and its second harmonic UV enables efficient laser ablation of polymethylmethacrylate [22], since the synthesized electric field has a larger instantaneous value than in single color cases. In Ref. [23], it is argued based on experiments and phenomenological modeling that the modification of sapphire under femtosecond IR laser and its third harmonics is enhanced by the contributions of various multiphoton absorption paths including not only

1. mzktni@atto.t.u-tokyo.ac.jp
2. corresponding author:ishiken@n.t.u-tokyo.ac.jp

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direct valence-conduction but also defect-level-mediated ones.

Photoionization in dielectrics under an intense laser pulse has been theoretically modeled by various methods including the Keldysh theory \cite{24,26}, rate equation \cite{27}, time-dependent Schrödinger equation within the independent electron approximation (IEA) \cite{28,29}, density matrix method \cite{30,34}, and time-dependent density functional theory (TDDFT) \cite{35,39}. By modeling fused silica, the Keldysh theory \cite{24,26}, rate equation \cite{27}, and time-dependent density functional theory (TDDFT) \cite{35,39}. By modeling fused silica, de Alaiza Martínez et al. \cite{40} have recently claimed that increase in energy absorption under simultaneous femtosecond IR and UV pulse irradiation is due to the interplay of various multiphoton ionization paths. This argument has been supported by Duchateau et al. \cite{41} also for a band-dispersion system, α-quartz. Although electron dynamics and high-harmonic generation has been extensively discussed in terms of the coupling between intraband and interband transitions for single-color cases \cite{29,31,33,42,49}, the role of the coupling under simultaneous two-color irradiation has been little studied. Thus, while increase in energy absorption and ablation efficiency by dual-color pulse irradiation appears to be quite general observation, a consensus has not been reached on its mechanisms, which possibly depend on materials and irradiation conditions.

In the present study, we investigate the energy absorption by bulk silicon under simultaneous dual-color (UV and IR) femtosecond laser fields, using numerical simulations based on the time-dependent density functional theory (TDDFT). TDDFT is an ab initio framework successfully applied to ultrafast carrier dynamics under intense laser-matter interaction \cite{33,50,57}. We employ the SALMON code \cite{58} and examine the dependence of energy absorption on mixing ratio $\eta$ of the two color components with the total intensity (or equivalently, fluence and energy) conserved. Our calculations show that the absorbed energy is significantly enhanced by dual-color irradiation and maximized at $\eta \sim 0.5$. Our analyses reveal that the intraband motion of the electrons driven in the valence band by the IR field has a substantial role in increasing valence-to-conduction interband transition induced by the UV field. These observations indicate that the strong-field electron excitation dynamics can be controlled by nonlinear coupling of a long-wavelength-driven intraband motion and a short-wavelength-driven interband transition.

This article is organized as follows. Section II describes our simulation methods. We briefly review TDDFT and describe how to evaluate absorbed energy. Section III shows the calculated ground state and linear response properties of Si. In Sec. IV we present and analyze our numerical results. Conclusions are given in Sec. V.

## II. TIME-DEPENDENT DENSITY FUNCTIONAL THEORY

The time propagation of an $N_e$-electron system under an optical field is calculated by solving the time-dependent Kohn-Sham (TDKS) equations for the Kohn-Sham orbitals $\{\psi_i(\mathbf{r}, t)\}$ \cite{59},

$$i\hbar \frac{\partial}{\partial t} \psi_i(\mathbf{r}, t) = -\frac{1}{2m} \nabla^2 \psi_i(\mathbf{r}, t) + V_{KS}[\psi_i(\mathbf{r}, t)] \psi_i(\mathbf{r}, t),$$  \hspace{1cm} (1)

where,

$$V_{KS}[\psi_i(\mathbf{r}, t)] = \frac{1}{2m} \left[ \mathbf{p} + e \mathbf{A}(t) \right]^2 + V_{\text{eff}}[\psi_i(\mathbf{r}, t)],$$  \hspace{1cm} (2)

denotes the velocity gauge Kohn-Sham Hamiltonian within the electric dipole approximation, $\mathbf{p}$ the canonical momentum, $e$ the elementary charge, $\mathbf{A}(t)$ the vector potential, $V_{\text{eff}}$ the effective potential (see below), $m$ the electron mass. The time-dependent electron density $n_e(\mathbf{r}, t)$ is given by,

$$n_e(\mathbf{r}, t) = \sum_{i \in \text{occ}} |\psi_i(\mathbf{r}, t)|^2.$$  \hspace{1cm} (3)

The effective potential $V_{\text{eff}}$, $V_{\text{eff}}[n_e(\mathbf{r}, t)] = V_{\text{ion}}(\mathbf{r}) + V_{\text{H}}[n_e(\mathbf{r}, t)] + V_{xc}[n_e(\mathbf{r}, t)],$ (4) consists of the electron-ion potential $V_{\text{ion}}$ consists of the norm-conserving pseudopotential \cite{60}, the Hartree potential $V_{\text{H}}$, and the exchange-correlation potential $V_{xc}$. We employ the modified Becke-Johnson (mBJ) potential \cite{61,62} for $V_{xc}$.

We consider a two-color pulse whose vector potential $\mathbf{A}(t)$ is described by,

$$\mathbf{A}(t) = \mathbf{A}_1(t) + \mathbf{A}_2(t),$$  \hspace{1cm} (5)

$$\mathbf{A}_1(t) = -a_1 \cos^2 \left( \frac{\pi}{T} \left( t - \frac{T}{2} - t_{\text{delay}} \right) \right) \sin \left( \omega_1 \left( t - \frac{T}{2} - t_{\text{delay}} \right) \right) \left( t_{\text{delay}} \leq t \leq t_{\text{delay}} + T \right),$$  \hspace{1cm} (6)

$$\mathbf{A}_2(t) = -a_2 \cos^2 \left( \frac{\pi}{T} \left( t - \frac{T}{2} \right) \right) \sin \left( \omega_2 \left( t - \frac{T}{2} \right) \right) \left( 0 \leq t \leq T \right),$$  \hspace{1cm} (7)

where $a_1, a_2$ denote the amplitude and polarization vectors of each color, $T$ the foot-to-foot pulse width (corresponding to 0.36T in FWHM), $\omega_1, \omega_2$ the central frequency of each color, and $t_{\text{delay}}$ the time delay between the two colors. $\mathbf{A}_1, \mathbf{A}_2$ are zero vectors on outside the time domain of definition ($t < t_{\text{delay}}$ or $t > T + t_{\text{delay}}$ for $\mathbf{A}_1$, $t < 0$ or $t > T$ for $\mathbf{A}_2$). We set $t_{\text{delay}} = 0$ unless explicitly stated.

We evaluate the absorbed energy as work done by the electric field $\mathbf{E}(t) = -\mathbf{A}(t)$,

$$W = -e \int_0^T d\tau \mathbf{J}(\tau) \cdot \mathbf{E}(\tau),$$  \hspace{1cm} (8)
with the current density,
\[ J(t) = \frac{2}{\hbar \Omega} \int_\Omega dr \sum_i \phi_i^*(r, t) [\hbar \mathbf{KS}, r] \phi_i(r, t), \]
where 0 is the volume of the simulation box.

Using the open source package SALMON, our simulation is conducted on a crystalline silicon primitive cell (see Appendix A for the crystal structure and the calculated band structure), which is discretized into 16 real-space and 16^3 k-space grids. The lattice constant a is set to 5.468 Å (the experimental value is 5.43 Å). The time step is 0.03 a.u., equivalent to 0.7257 as. These parameters are finer than those used in the previous works [37, 58]. We have also confirmed the convergence of simulation results (see also Appendix B). The laser polarization is assumed to be parallel to ΓX direction unless specified otherwise.

III. GROUND STATE AND LINEAR RESPONSE

The initial ground state of the system is taken as the eigen state of the field-free Hamiltonian \( h_{\mathbf{KS}} | \mathbf{A} = 0 \rangle \). Figure 1 shows the density of states of the system. The indirect bandgap is found to be perfectly agreeing with the experimental one, 3.1 eV [64].

The direct bandgap is estimated by the imaginary part of the dielectric function, obtained by time propagation after an impulsive momentum kick. This is realized by a step-like vector potential,
\[ \mathbf{A}(t) = \begin{cases} -\mathbf{A}_0 & (t \geq 0) \\ 0 & (t < 0) \end{cases}, \]
which corresponds to an impulsive electric field,
\[ \mathbf{E}(t) = \mathbf{A}_0 \delta(t). \]

Noting that the electric field has a constant power spectrum over all frequencies, the diagonal component of the optical conductivity is calculated as,
\[ \sigma_m(\omega) = -\frac{\epsilon \hat{J}_m(\omega)}{A_{0m}} \quad (m = x, y, z), \]

where \( \hat{J}_m \) denotes the m component of the temporal Fourier transform of the current density. Assuming isotropic media, the dielectric function \( \varepsilon_m(\omega) \) is given by,
\[ \varepsilon_m(\omega) = 1 + 4\pi i \sigma_m(\omega) / \omega. \]

The imaginary part of thus evaluated dielectric function is consistent with the measured one [65] (Fig. 2). The direct bandgap is found to be 3.1 eV, which quasi-perfectly agrees with the experimental one, 3.2 eV [66].

IV. RESULTS AND DISCUSSIONS

A. Energy transfer from laser to electrons

Let us consider energy transfer to silicon from superposed two-color fields with \( h_{\omega_1} \) and \( h_{\omega_2} \) photon energies \( (\hbar \omega_1 \leq \hbar \omega_2) \) and both 14.4 fs FWHM pulse width. Whereas this pulse width is short compared to those typically used in experiments, computationally demanding TDDFT simulations for such short pulses have been useful to investigate fundamental laser-solid interactions [67, 59, 41]. We examine how the absorbed energy varies with mixing ratio \( \eta = I_1/(I_1 + I_2) \) with \( I_i \) (i = 1, 2) being the peak intensity of color component i while fixing the total intensity \( I_{\text{tot}} = I_1 + I_2 \). Figure 3 displays the
temporal profiles of the electric field, the current density, and the absorbed energy for $I_{\text{tot}} = 10^{12}$ W/cm$^2$. The orange and cyan solid lines are for the single-color ($\eta = 0$) and dual-color ($\eta = \frac{1}{2}$) fields, respectively.

The absorbed energy is significantly (ca. three times) higher for the two-color case. If we naively assumed an incoherent sum of two-photon excitation by $\hbar \omega_1$ and single-photon ionization by $\hbar \omega_2$, the absorbed energy would scale as $\sigma_1 \eta^2 + \sigma_2 (1 - \eta)$ with $\sigma_1 (\sigma_2)$ being excitation cross section by $\hbar \omega_1 (\hbar \omega_2)$, from which we would expect decrease, rather than increase, in absorbed energy by two-color mixing. Indeed, if the two pulses are separated in time, energy absorption is significantly reduced and even smaller than in the single-color case (Fig. 3). Thus, simultaneous, rather than consecutive, irradiation is essential to the enhancement.

Let us further examine the dependence of the absorbed energy on mixing ratio $\eta$, photon energies $\hbar \omega_{1,2}$, laser polarization orientation, and intensity $I_{\text{tot}}$. Figure 4 shows the results as a function of $\eta$ for $I_{\text{tot}} = 10^{12}$ W/cm$^2$, $\hbar \omega_2 = 3.2$ eV (387.5 nm wavelength) and three different values of $\hbar \omega_1$. Note that $\hbar \omega_2$ (the shorter-wavelength component) is larger than the direct band gap. The green stars in Fig. 5 indicate the transferred energy from the laser field whose polarization is 45° to the $\Gamma X$ direction, they exhibit almost same trend with the original laser polarization. Again, combining two colors significantly enhances the energy transfer, maximized at $\eta \sim \frac{1}{2}$. It should be noticed that the enhancement effect is larger with smaller $\hbar \omega_1$, or equivalently, longer wavelength of the first color, in contrast to the fact that, for $\eta = 1$, the absorbed energy is smaller with smaller $\hbar \omega_1$. Since the longer the wavelength, the larger the vector-potential amplitude and, thus, the crystal-momentum shift of elec-
Indeed, the Keldysh parameter for \( \hbar \omega_1 \) and \( \hbar \omega_2 = 3.2 \) eV indicated in the legend. Green stars: both laser polarization of \( \hbar \omega_1 = 1.6 \) eV and \( \hbar \omega_2 = 3.2 \) eV are rotated \( \pi/4 \). Brown crosses: delayed irradiation (\( t_{\text{delay}} = T \)) of \( \hbar \omega_1 = 0.2 \) eV and \( \hbar \omega_2 = 3.2 \) eV. Inset: close-up of the results for \( \eta = 1 \).

The absorbed energy is nearly independent of relative phase \( \eta \) for \( \hbar \omega_1 = 0.2 \) eV (6.2 \( \mu \)m wavelength) and three different values of \( \hbar \omega_2 \), we can see larger absorption for larger \( \hbar \omega_2 \) (the shorter-wavelength component), which coincides with the behavior of linear absorption between 2.8 and 4.0 eV (Fig. 2). This observation suggests that the major role of \( \hbar \omega_2 \) is to induce interband excitation.

In Fig. 8 we display the absorbed energy normalized to the case \( \eta = 0 \) [\( W(\eta)/W(\eta = 0) \)] as functions of mixing ratio \( \eta \) and total peak intensity \( (I_1 + I_2 = 10^{12} - 10^{13} \text{ W/cm}^2) \) for \( \hbar \omega_1 = 0.2 \) eV (6200 nm wavelength) and \( \hbar \omega_2 = 3.2 \) eV (387.5 nm).

The absorbed energy is nearly independent of relative phase between the two color fields (Fig. 8). This observation suggests that tunneling ionization is not dominant in the present situation. Indeed, the Keldysh parameter for 1.6 (3.2) eV photon energy and \( 10^{12} \text{ W/cm}^2 \) intensity is 5.3 (10), which corresponds to the multi-photon regime.

In Fig. 7 which plots the absorbed energy vs. \( \eta \) for \( \hbar \omega_1 = 0.2 \) eV (6.2 \( \mu \)m wavelength) and three different
energy absorption. Then, is it increase in the former or the latter that accounts for the enhanced energy deposition enhancement found in the previous subsection? To reveal the origin of the energy deposition enhancement under double-color laser fields, we address to $\eta$, $\hbar \omega_1$, and $\hbar \omega_2$ dependence of these two quantities. The number of excited electrons $n_{ex}$ is evaluated as,

$$n_{ex} = \sum_{i \in \text{cond}} 2 \left| \int \! dr \sum_j \phi_i^*(r, 0) \phi_j(r, T + t_{\text{delay}}) \right|^2,$$

and the absorbed energy per carrier $E_{\text{mean}}$ as,

$$E_{\text{mean}} = \frac{W}{n_{ex}}.$$  \hfill (15)

Figure 9 shows $n_{ex}$ and $E_{\text{mean}}$ as functions of the mixing ratio $\eta$ for $\hbar \omega_2 = 3.2$ eV and three different values of $\hbar \omega_1$. We can see that $n_{ex}$ follows the trend in Fig. 4 strongly enhanced by dual-color irradiation, compared with the single-color cases ($\eta = 0.1$), and peaking at $\eta \sim 0.5$ [Fig. 9a]. The enhancement of interband excitation is larger for smaller $\hbar \omega_1$, again consistent with the trend in Fig. 4. On the other hand, the absorbed energy per carrier $E_{\text{mean}}$ is surprisingly nearly independent of $\eta$ for $\hbar \omega_1 = 0.8$ and 1.6 eV [Fig. 9b]. Interestingly, $E_{\text{mean}}$ is larger than the direct bandgap (3.1 eV), because not only the lowest but also higher conduction bands are populated (see Appendix C). For the case of $\hbar \omega_1 = 0.2$ eV, although $E_{\text{mean}}$ gradually increases with $\eta$, it reaches maximum at $\eta \sim 0.9$, rather than $\sim 0.5$. Furthermore, the enhancement factor (two at most) is much smaller than that of $W$ and $n_{ex}$.

Figure 10 displays $n_{ex}$ [panel (a)] and $E_{\text{mean}}$ vs. $\eta$ [panel (b)] shows $n_{ex}$ for $\hbar \omega_1 = 0.2$ eV and three different values of $\hbar \omega_2$. Again, the number of excited electrons $n_{ex}$ follows a trend similar to that in Fig. 7, peaking at comparable mixing of the two colors and increasing with $\hbar \omega_2$. In contrast, the mean absorbed energy $E_{\text{mean}}$ is nearly independent of $\hbar \omega_2$ and only a weak function of $\eta$.

Hence, the energy absorption enhanced by two-color pulse irradiation originates mainly from increase in excitation from the valence to the conduction band, rather than further excitation of carriers generated in the conduction band. These findings suggest the following mechanism underlying the increased energy absorption under two-color laser fields. The shorter-wavelength component $\hbar \omega_2$, if alone, would excite only valence electrons of crystal momenta $k_{\text{res}}$ where $\hbar \omega_2$ is resonant with the excitation energy. In two-color cases, however, the longer-wavelength component $\hbar \omega_1$ drives the intraband motion in the valence band; the electronic momentum is shifted as $\mathbf{k}(t) = \mathbf{k}_0 + \mathbf{A}_1(t)$, with $\mathbf{k}_0$ being its field-free value. The peak amplitude of the vector potential $\mathbf{A}_1(t)$ is 0.06451, 0.1286, and 0.5145 atomic units (a.u.) for $\hbar \omega_1 = 1.6$, 0.8, 0.2 eV, respectively,
at $I_1 = 5 \times 10^{11}$ W/cm$^2$. These values are to be compared with the half width of the first Brillouin zone along $\Gamma X$, 0.304 a.u. Thus, a substantial part of valence electrons can pass by the resonant momenta $k_{\text{res}}$ during the intraband motion, enabling much more electrons get excited. From the viewpoint of $\hbar \omega_1$, on the other hand, it is much smaller than the band gap, so that excitation is not much induced by the longer-wavelength component alone but facilitated by mixing the shorter-wavelength component $\hbar \omega_2$. The interplay and balance between the intraband motion by $\hbar \omega_1$ (through vector potential) and interband excitation by $\hbar \omega_2$ (through photon energy) leads to increase in generated carriers, as in Figs. 9(a) and 10(a), and to maximum enhancement of energy absorption at approximately equal two-color mixing. Interestingly, for the case of $\hbar \omega_1 = 0.2$ eV, the maximum momentum shift (0.5145 a.u.) given by the vector potential amplitude exceeds the Brillouin zone radius. Hence, excited electrons can be shifted in the conduction to reach the Brillouin zone edge and then further excited to upper bands, which results in increased mean absorbed energy [Figs. 9(b) and 10(b)]. It should be emphasized, nevertheless, that the intraband motion in the valence band plays a dominant role in the enhanced energy absorption, rather than that in the conduction band.

C. Population analysis in $k$-space

To further verify the above-proposed mechanism, we analyze the time-dependent $k$-resolved population defined by,

$$\rho_{k,i}(t) = \left( \int dr \sum_{j \in \text{occ}} \phi_{k+e\mathbf{A}(t),i}^* (r,0) \phi_{k,j} (r,t) \right)^2,$$  \hspace{1cm} (16)

where $i$ and $j$ denote the band index. We consider photon energy combination $\hbar \omega_1 = 1.6$ eV and $\hbar \omega_2 = 3.2$ eV ($I_{\text{tot}} = 10^{12}$ W/cm$^2$) and focus on the following three $k$-points: $\Gamma$ ($k = [-0.019, -0.019, -0.019]$), $\Gamma_1$ ($k = [0.019, -0.019, 0.057]$), and $P$ ($k = [-0.019, -0.019, -0.095]$).

For the case of $\eta = 0.0$ [Fig. 11(a)], the population at and near the $\Gamma$ point ($\Gamma, \Gamma_1$) exhibits Rabi flopping [45, 67], which means the excitation around the $\Gamma$ point is saturated, while almost no electrons are excited at the $P$ point, which is far from the $\Gamma$ point. Thus, the energy transfer takes place only around the $\Gamma$ point, and even there, the laser pulse energy is “wasted”. For $\eta = 0.5$ [Fig. 11(b)], on the other hand, while the conduction band is still populated at $\Gamma$ and $\Gamma_1$ as much as for $\eta = 0$, the $P$ point electron is comparably excited. This observation indeed supports our discussion in the previous subsection that mixing the lower photon energy component into the driving laser field induces intraband motion in the valence band, expressed by subscript $k + e\mathbf{A}(t)$ in Eq. (16), and broadens the excitable crystal moment range, leading to more efficient use of the laser energy. Whereas we also see increase in residual population even at the $\Gamma$ point, it happens accidentally during the Rabi oscillation. Indeed, under a shorter pulse (7.2 fs in FWHM, $I_{\text{tot}} = 10^{12}$ W/cm$^2$), the final population of the $\Gamma$ and $\Gamma_1$ points for $\eta = 0.5$ [Fig. 12(b)] is smaller than for $\eta = 0$ [Fig. 12(a)]. Nevertheless, the absorbed energy is still maximized at $\eta \sim 0.5$ [Fig. 12(c)]. In Fig. 13, we compare the residual population distributions in the conduction band shown as a sectional view along $k_x = -0.019$ a.u. for $\eta = 0.0$ and $\eta = 0.5$ after the short laser pulse (7.2 fs in FWHM, $I_{\text{tot}} = 10^{12}$ W/cm$^2$) irradiation. Again, we find that wider area of the $k$ space is excited for $\eta = 0.5$ [Fig. 13(b)] than for $\eta = 0.0$ [Fig. 13(a)], leading to higher total excitation probability. These observations indicate that not the population transfer at the $\Gamma$ point alone but the excitable $k$ range expanded by the addition of the longer-wavelength component has a major contribution to the enhanced energy absorption.

V. CONCLUSIONS

We have investigated energy absorption by crystalline silicon under two-color laser pulse irradiation, by varying the mixing ratio with the total intensity or, equivalently, fluence fixed constant. We have specifically considered the higher photon energy $\hbar \omega_2$ (3.2 eV) in the UV
range above the optical gap energy $E_g$ and the lower one $\hbar \omega_1$ (0.2-1.6 eV) in the infrared below it. Our TDDFT computation have shown that energy transfer is substantially enhanced by simultaneous two-color irradiation and maximized when both color components are roughly equally mixed. The longer the wavelength of the longer-wavelength component, the more significant the effect. Increase in carrier generation, rather than that in absorbed energy per carrier, accounts for increase in the total absorbed energy. All these observations, along with our $k$-resolved population analysis, have revealed that the electron dynamics in the valence band driven by the longer-wavelength component $\hbar \omega_1$ plays a crucial role, in contrast to previous studies, which have emphasized the interplay of different multiphoton paths or the dynamics of the generated carriers. The $\hbar \omega_2$ field induces interband excitation (carrier injection) around the $\Gamma$ point, where the transition energy is resonant with $\hbar \omega_2$. On the other hand, the $\hbar \omega_1$ field of a large vector potential amplitude (corresponding to the maximum momentum shift) drives intraband motion in the valence band, enabling electrons initially far from the $\Gamma$ point to pass through the resonance, thus, extending the excitable $k$ range. Additionally, for the case of $\hbar \omega_1 = 0.2$ eV, the induced momentum shift exceeds the width of Brillouin zone along $\Gamma X$ and promotes further excitation of carrier electrons to upper conduction bands.

The decoherence would have some effects depending on dephasing time $T_2$. Floss et al. [68] have pointed out that $T_2$ is order of 10 femtoseconds, which is longer than the time scale of Rabi oscillation observed in our calculation. Freeman et al. [69] have recently shown that relaxation effect on high-harmonic generation emerges only after several tens of femtoseconds. Hence, our simulation results are semi-quantitatively valid even in the presence of the generated carriers.
of the decoherence effect.

The degree of ionization and energy per electron are rather high for the photon energy combination \(0.2 \text{ eV}, 3.2 \text{ eV}\). Thus, we should be cautious if we discussed phenomena at longer time scale, which eventually leads to material processing. On the other hand, in this work, we limit ourselves to the ultrashort time scale before the lattice starts to move significantly. Moreover, the photon energy combination \(1.6 \text{ eV}, 3.2 \text{ eV}\), for which the degree of ionization is \(4\%\), is used in the main part of our analysis. The Rabi oscillation observed in Fig. 11 implies that the bandgap is close to the original one, thus, indicates that our discussion of the ultrafast electron excitation dynamics based on the ground state band structure is approximately valid and useful for physical understanding of the early stage of laser material processing.

The findings of this work, suggesting dramatic improvement of femtosecond laser ablation rate of bandgap materials, could be experimentally examined, using the combination of ultrashort intense mid-infrared (MIR) and ultraviolet laser pulses. Thereby, increase in generated carrier will be easier to probe directly rather than absorbed energy itself. Use of a terahertz pulse instead of IR may be even more advantageous, since its vector potential has an even larger amplitude.

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Appendix A: Crystal structure and band structure

Figure 14 shows the Wigner-Seitz cell of the crystalline silicon in reciprocal space. The image is generated using XCrySDen [72].

![Fig. 15. Energy dispersion for the conduction and valence bands of silicon, calculated using ABINIT code [70, 71] with the lattice constant \(a = 5.468 \text{ Å}\). Red and blue lines indicate conduction and valence bands respectively.](image)

Appendix B: Convergence with respect to grid spacings and time step

Figure 16 compares the imaginary part of the dielectric function \(\text{Im}[\varepsilon]\) calculated with two different real(\(k\))-space grid widths and time steps. Red dashed line indicates \(\text{Im}[\varepsilon]\) with 16\(^3\) grid points in real-space and \(k\)-space, blue line with 18\(^3\) grid points in real-space and 24\(^3\) grid points in \(k\)-space. The time step is 0.03 a.u. with the former parameter set, 0.02 a.u. with the latter. Both results are perfectly overlapped, indicating the simulation re-
FIG. 16. Imaginary part of the dielectric function using our parameters (light blue solid line) and finer parameters (red dashed line) for real-space, $k$-space, and time discretization. See text for the parameter values.

Appendix C: Excited carrier population in each conduction band

Figures [17,20] show the residual population map of each conduction band within the same cross section as Fig. [18] for $\eta = 0.5$ under $\hbar \omega_1 = 1.6 \text{ eV}$ and $\hbar \omega_2 = 3.2 \text{ eV}$ ($I_1 + I_2 = 10^{12} \text{W/cm}^2$) and the energy gap between the valence top band and each conduction band. We see that the electrons are excited around the minimum bandgap (3.1 eV) but also at $k$ points where the energy gap is larger than it. Therefore, the mean absorbed energy exceeds the direct bandgap (3.1 eV) [see Fig. [8b]].

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FIG. 17. (a) The residual population of the lowest conduction band on a cross section ($k_x = -0.019$ a.u.) after irradiated by the short pulse (7.2 fs in FWHM, $I_{\text{tot}} = 10^{12}$ W/cm$^2$, $\eta = 0.5$). (b) Energy gap between the lowest conduction band and the highest valence band. Negative value is assigned outside the first Brillouin zone.

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FIG. 20. (a) The residual population of the 4th lowest conduction band on a cross section \((k_z = -0.019 \text{ a.u.})\) after irradiated by the short pulse (7.2 fs in FWHM, \(I_{\text{tot}} = 10^{12} \text{ W/cm}^2, \eta = 0.5\)). (b) Energy gap between the 4th lowest conduction band and the highest valence band. Negative value is assigned outside the first Brillouin zone.

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