Photoinduced nonequilibrium electron dynamics in solids have attracted much attention of researchers not only in condensed matter physics but also in optical physics. Recent great progress in intense laser pulse, ultrafast time-resolved experimental probes, and theoretical methods for nonequilibrium systems has opened up a new research field of exotic light-induced phenomena. High-order harmonic generation (HHG) is one of the attractive phenomena induced by intense laser light \([1, 2]\). This is a nonlinear and nonperturbative light-matter coupled phenomenon, and is widely recognized to be utilized to generate the attosecond X-ray laser pulse. Studies of HHG have been developed in atom- and molecule-gas systems \([3, 4]\), in which the HHG spectra consist of a characteristic plateau and cut-off energy. This behavior is well explained by the three-step model, i.e., a sequential process of ionization, forced oscillatory motion, and recombination of electrons in atomic/molecular potentials \([7, 8]\). In crystalline solids where the atom are aligned periodically, electronic processes involved in HHG are considered on the basis of the one-body Bloch-band picture is applicable, correlated electron systems have great potentialities of HHG. Large energy-band structures arising from electron itineracy \([1–3, 10–24]\).

In this Letter, we show that HHG spectra emerge owing to many-electron dynamics in a correlated electron system, rather than the Bloch electron itineracy. We analyze photoinduced dynamics of an interacting fermion model on a dimer-type lattice and its low-energy effective model described by the pseudo-spin (PS) operators. We find emergence of HHG spectra in a spontaneously symmetry-broken state, in which charge densities are polarized inside of dimer units, show threshold behavior with respect to light amplitude and are interpreted in terms of tunneling and recombination of kink-antikink excitations in an electric field.

High-Harmonic Generation in a Correlated Electron System

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High-harmonic generation (HHG) in crystalline solids have been examined so far on the basis of one-body energy-band structures arising from electron itineracy in a periodic potential. Here, we show emergence of HHG signals which are attributed to dynamics of many-body states in a low-dimensional correlated electron system. An interacting fermion model and its effective pseudo-spin model on a one-dimensional dimer-type lattice are analyzed. Observed HHG signals in a spontaneously symmetry-broken state, where charge densities are polarized inside of dimer units, show threshold behavior with respect to light amplitude and are interpreted in terms of tunneling and recombination of kink-antikink excitations in an electric field.

The HHG spectra are polarized inside of dimer units. The HHG spectra are attributed to dynamics of many-body states in a low-dimensional correlated electron system. An interacting fermion model and its effective pseudo-spin model on a one-dimensional dimer-type lattice are analyzed. Observed HHG signals in a spontaneously symmetry-broken state, where charge densities are polarized inside of dimer units, show threshold behavior with respect to light amplitude and are interpreted in terms of tunneling and recombination of kink-antikink excitations in an electric field.

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inter-dimer Coulomb interaction and the intra-dimer hopping in Eq. (1), respectively. This model is suitable to study the collective excitations, i.e., the kink and antikink (domain-wall) excitations.

A vector potential of light is introduced as the Peierls phase as $t_0 \rightarrow t_0 e^{-i A(t)}$ and $t' \rightarrow t' e^{-i A(t')}$ in Eq. (1), where $A(t)$ is the vector potential at time $t$ and the difference in the bond lengths are neglected. The electric field is given by $E(t) = -\partial A(t)/\partial t$. This coupling corresponds to the rotation of the transverse field as follows $\mathcal{H}_F$ is replaced by $-\hbar/2 \sum_i \left[ \cos A(t) \sigma^i_z - \sin A(t) \sigma^i_y \right]$, and the electric current operator is identified as $\hat{j}(t) = -(i/eN) \sum_i \left[ \sin A(t) \sigma^i_z + \cos A(t) \sigma^i_y \right]$. We confirmed that the numerical results of HHG in the above two models qualitatively coincide in the polar CO state, and we will mainly present the results for the TI model.

The GS and excited states in the TI model without the light field have been settled $\psi_G$ and $|\psi^+_G\rangle$. The GS is the DM insulating state (a paramagnetic PS state), i.e., $\langle \sigma^z \rangle = 0$ for $V'/(4t_0) < 1$, and is the polar CO state (a ferromagnetic PS state) with spontaneously breaking of the space-inversion symmetry, i.e., $\langle \sigma^z \rangle \neq 0$ for $V'/(4t_0) > 1$. The boundary at $V'/(4t_0) = 1$ is the quantum critical point. In order to calculate the transient dynamics induced by the light field in the thermodynamic limit, the infinite time-evolving block decimation (iTEBD) method is adopted $\mathcal{H}(\tilde{t})|\psi^+_G\rangle$ with a small time difference $\delta \tilde{t}$ and the time-dependent Hamiltonian $\mathcal{H}(\tilde{t})$. In most of the numerical calculations, the maximum number of the matrix dimension $\chi$ in the iTEBD method, and the time difference are chosen to $\chi = 200$ and $\delta \tilde{t} = 0.01/\hbar_0$, respectively, which are enough to obtain well convergent results as shown later (see inset of Fig. 1(c)). We also adopt the exact diagonalization (ED) method based on the Lanczos algorithm for finite size clusters, where the total number of dimer units is $N = 16$ and 18 with the periodic-boundary condition. The optical absorption spectra [see Fig. S.4 in the Supplemental Material (SM)] is schematically depicted in Fig. 1(b). In the polar CO state, the excitation spectra are attributed to the kink-antikink pair excitations, and exhibit a continuous band where the upper and lower edges of the band are $E_H = 4(V'/4 + t_0)$ and $E_L = 4(V'/4 - t_0)$, respectively. In the DM state, the low-energy collective excitation is located at $2(t_0 - V'/4)$.

First, we show the HHG spectra in the polar CO state $|V'/4(t_0) = 2.4]\rangle$ in the continuous-wave (cw) light. We set $A(t) = -A_0 e^{-t^2/(2\tau)^2} \cos(\omega_0 t)$ for $t < 0$, and $A(t) = -A_0 \cos(\omega_0 t)$ for $t > 0$ with frequency $\omega_0$, amplitude $A_0$, and raising time $\tau$. Numerical values of $\omega_0$ are chosen to be much smaller than the excitation energy gap $\Delta_{\text{gap}} = E_L$. Time profiles of the electric current $\langle \hat{j}_t \rangle$ and its Fourier transform $\langle \hat{j}_{\omega_0} \rangle$ as well as $A(t)$, are shown in Figs. 1(d) and 1(e). A multiple pulse-like profile with period of $T_0 \equiv 2\pi/\omega_0$ appears in $\langle \hat{j}_t \rangle$, and a series of sharp spikes at $\omega = n\omega_0$ with integer number $n$ appear in $\langle \hat{j}_{\omega_0} \rangle$ [see inset of Fig. 1(e)]. The HHG spectra show a plateau approximately between $E_L$ and $E_H$, indicating the nonperturbative processes for this HHG. Owing to the breaking of the space-inversion symmetry in the GS, both the odd and even orders of high harmonics emerge. Overall features mentioned above do not depend on $\chi (\geq 100)$ and are almost reproduced by the ED method in finite-size clusters as shown in Fig. S.2 in SM.

The HHG spectra are sensitive to the light amplitude $A_0$. In Figs. 2(a) and 2(b), the intensity map of $\langle \hat{j}_{\omega_0} \rangle$ in the $\omega - A_0$ plane and its enlargement are shown, respectively. The threshold behavior of the HHG spectra with respect to $A_0$ is clearly seen. We find that the threshold decreases with decreasing $V'$ (see Fig. S.1 in SM).

The observed HHG is understood by repetition of dynamics induced by a one-cycle pulse. Thus, to reveal the threshold behavior in more detail, we examine responses to a one-cycle pulse given by $A(t) = -A_0 e^{-t^2/(2\tau)^2} \cos(\omega_0 t)$. Using the iTEBD method, we analyze the absorbed energy defined by $\Delta E \equiv \mathcal{E}(t \gg \tau) - \mathcal{E}(t \ll -\tau)$ with total energy $\mathcal{E} = \langle \mathcal{H}_G \rangle/N$, which reflects population of the excited states induced by the pulse field. In Fig. 2(c), we plot $\Delta E$ as a function of $1/E_0$ with the electric field amplitude $E_0 \equiv A_0\hbar_0$. We note that, instead of the vector potential, a response to the electric field is suitable to examine the breakdown phenomena which will be introduced later. The current operator in this case is defined $\langle H(t)\rangle$.
in Eq. (8) in SM. The exponential dependence observed as \( \Delta E \propto \exp(-E_0/|E_0|) \) with a threshold electric field \( E_0 \). A deviation of data from this function for \( \Delta E/|E_0| < 10^{-9} \) is attributed to the numerical artifact. This behavior implies a nonperturbative process in HHG, and is reproduced by the ED method in finite clusters [see Fig. S.3(c) in SM]. The threshold amplitude calculated in several values of \( V \) is scaled by the excitation energy gap \( \Delta_\text{gap} = 4V/4 - t_0 \) as \( E_\text{th}/\Delta_\text{gap} \) with \( \alpha \sim 1.59 \) as shown in Fig. 2(d). This indicates a Landau-Zener-like breakdown in the HHG, except for the exponent which is different from \( \alpha = 2 \) in a static field.

The observed HHG spectra and their characteristic time profiles are interpreted through the following analysis based on adiabatic kink-antikink dynamics. We consider the TI model in an electric field, \( \mathcal{H}_\text{TI} = -[E(t)/2] \sum_i \sigma_i^z \), and exhaust this by using the ED method. The energy levels as functions of a static electric field \( E(t) = E_s \) is shown in Fig. 3(a). The eigen wavefunction and eigen energy for finite \( E_s \) are denoted by \( |\phi_i(E_s)\rangle \) and \( E_i(E_s) \) (\( i \geq 0 \)), respectively, which are adiabatically connected to the \( i \)th eigen state at \( E_s = 0 \). The GS at \( E_s = 0 \) are doubly degenerated in the thermodynamic limit, i.e., the all-up and all-down states, schematically \( \cdots \uparrow \uparrow \uparrow \downarrow \downarrow \cdots \) and \( \cdots \downarrow \downarrow \downarrow \uparrow \uparrow \cdots \), respectively, and the excited states are continuum with the finite excitation gap from GS. With increasing \( E_s \) from zero, the energy of the all-up (all-down) state decreases (increases).

Then, we examine the current induced by the cw field shown in Fig. 3(c) and Fig. 3(c) (red line) from a viewpoint of the adiabatic dynamics of many-body states. The wavefunction at time \( t \) is expanded as \( |\psi(t)\rangle = \sum_{i \geq 0} c_i |\phi_i(E_s)\rangle \exp[-iE_i(E_s)\tau] \) with coefficients \( c_i = \langle \phi_i(E_s)|\psi(0)\rangle \). Here, we assume that \( E_s = 0 \) is equal to \( E(t) \) and \( |\phi_0(E_s)\rangle \) is adiabatically connected to the all-up state at \( E_s = 0 \). Since \( |c_0|^2 \approx 1 \) and \( |c_{z\pm}|^2 \ll 1 \) as shown in Fig. 3(d), the current at time \( t \) is approximately given by \( \langle j \rangle_t = \langle \psi(t)|j|\psi(t)\rangle \approx \sum_{i \geq 0} c_i^* c_i \langle \phi_i(E_s)|\hat{J}|\phi_i(E_s)\rangle \exp[-i(E_i(E_s) - E_0(E_s))\tau] + \text{c.c.} \). In Fig. 3(c), we compare a time profile of the current calculated by the above method shown by blue line with that by the real-time evolution. We adopt the most dominant excited state among \( |\phi_i(E_s)\rangle \)'s. The two results almost coincide. We conclude that this picture based on the adiabatic dynamics is valid to understand the real time processes in the present HHG. The facts \( |c_0|^2 \approx 1 \) and \( |c_{z\pm}|^2 \ll 1 \) reflect the off-resonant excitation with the light frequency \( \omega_0 = \Delta_\text{gap} \) and nonperturbative tunneling processes are incorporated in \( c_i \) for \( i \geq 1 \). In the time profile of \( \langle j \rangle_t \), in Fig. 3(c), a fine oscillation is attributed to the exponential factor \( \exp[-i(E_i(E_s) - E_0(E_s))\tau] \). An envelope with period of \( T_0 = \frac{2\pi}{\omega_0} \), showing large amplitude in regions with positive \( E(t) \) (shaded areas in Fig. 3(c)), is due to the amplitude factor \( c_0 \langle \phi_0(E_s)|\hat{J}|\phi_i(E_s)\rangle \). As shown in Fig. 3(c), this characteristic time profile of the amplitude factor does not originate mainly from the populations of the excited state \( |c_{i\pm}|^2 \), but the transition amplitude \( |\langle \phi_0(E_s)|\hat{J}|\phi_{i\pm}(E_s)\rangle| \).

![Fig. 2](image1.png)

![Fig. 3](image2.png)
The green (dark gray) line in lower represents $\langle \hat{J}_\omega \rangle$ shown in Fig. 4(b). Amplitude of the electric current is dominated by the intra-dimer component, and contribution from the inter-dimer current is less than 10% (see green line in Fig. 4(a)), although amplitude of $\tau'$ is chosen to be half of $t_0$. We conclude that the essential characters in the HHG in the polar CO state is not governed by the electron propagation over the dimer units, but by the kind-antikink excitations and propagations.

In summary, we investigated HHG in the spontaneously symmetry-broken state realized in the TI model and the spinless-fermion model as the effective models of the interacting electrons in a dimer-type lattice structure. The kink-antikink dynamics are responsible for the present HHG, instead of the electron itineracy. The many-body character in the wavefunction governs the transition amplitude between GS and excited states. Experimental observations are crucial to confirm the present theoretical prediction for a new mechanism of HHG. Candidate materials are low-dimensional organic molecular solids, (TMTTF)$_2$X (TMTTF=Tetramethyltetraethylidifluorovalene, X=PF$_6$, AsF$_6$) which show the polar CO phase in low temperatures [41–43].

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In this section, we discuss the effective Hamiltonians of the interacting electron system in a dimer-type lattice structure. We start from the extended Hubbard model on a one-dimensional dimer-type lattice given by

$$
\mathcal{H} = -\sum_{is} \left( t_0 c_{i\sigma s}^\dagger c_{i+1\sigma s} + \text{H.c.} \right) - \sum_{is} (t' c_{i\sigma s}^\dagger c_{i+1\sigma s} + \text{H.c.}) + U \sum_i \bar{n}_{i\uparrow} \bar{n}_{i\downarrow} + V_0 \sum_i \bar{n}_{i\uparrow} \bar{n}_{i+1\uparrow},
$$

(S.1)

where $c_{i\sigma s}^\dagger$ ($c_{i\sigma s}$) is the creation (annihilation) operator of an electron at site $i$ with spin $s$ ($= \uparrow, \downarrow$) and sublattice $\gamma$ ($= a, b$) with spin $s$ ($= \uparrow, \downarrow$), and $\bar{n}_{i\uparrow} = \sum_s c_{i\sigma s}^\dagger c_{i\sigma s}$ is the number operator. The first two terms represent the electron hoppings between the nearest neighbor (NN) sites, the third term describes the on-site Coulomb interaction, and the last two terms describe the long-range Coulomb interactions between the NN sites. The electron number per site is set to 0.5.

In the limit of large $U$ in comparison with other energy parameters, it is expected that the electron occupancy in each site is less than one, and the spin degree of freedom does not play essential role in the optical processes. Then, the electron operators are replaced by the spinless-fermion operators, the third term in Eq. (S.1) is neglected, and thus the effective Hamiltonian is given by

$$
\mathcal{H}_{\text{SF}} = -\sum_{i} \left( t_0 f_{i\uparrow a}^\dagger f_{i\uparrow a} + \text{H.c.} \right) - \sum_{i} (t' f_{i\uparrow b}^\dagger f_{i+1\uparrow a} + \text{H.c.}) + V_0 \sum_i n_{i\uparrow} n_{i+1\uparrow} + V' \sum_i n_{i\uparrow} n_{i+1\uparrow},
$$

(S.2)

which is Eq. (1) in the main text (MT). We introduce the creation (annihilation) operator of a spinless fermion $f_{i\gamma}^\dagger$ ($f_{i\gamma}$) at the $i$th unit cell and sublattice $\gamma$, and the number operator for a spinless fermion $n_{i\gamma} = f_{i\gamma}^\dagger f_{i\gamma}$. The number of the spinless fermion per site is 0.5.

In the limit of the strong dimerization, i.e., $t' \ll t_0, V_0$, it is convenient to describe the electronic states inside a dimer unit by using the pseudo-spin (PS) operator with magnitude of 1/2. We introduce the PS operator as

$$
\sigma_i = \sum_{\gamma\gamma'} f_{i\gamma}^\dagger \bar{\sigma}_{\gamma\gamma'} f_{i\gamma'},
$$

(S.3)

with the Pauli matrices $\bar{\sigma}$. The intra-dimer Coulomb interaction term is replaced by a constant, and the inter-dimer one is transformed into $-V'/4 \sum_i \sigma_i^z \sigma_{i+1}^z$. Although additional inter-dimer interactions are induced by the second order processes with respect to $t'$, we neglect these terms for simplicity. Then, we have the transverse Ising (TI) model as an effective model of Eq. (S.2) given as

$$
\mathcal{H}_{\text{TI}} = -\frac{V'}{4} \sum_i \sigma_i^z \sigma_{i+1}^z - t_0 \sum_i \sigma_i^x,
$$

(S.4)

which is Eq. (2) in MT.

In the light field, the vector potential $A(t)$ is introduced in Eq. (S.2) as the Peierls phase as $t_0 \rightarrow t_0 e^{iAt}$ and $t' \rightarrow t' e^{-iAt}$, where a difference of the bond lengths is neglected. In the TI model, in which the linear term of $t'$ is neglected, the coupling with the vector potential is introduced by replacing the second term in Eq. (S.4) as

$$
- t_0 \sum_i \left[ \cos A(t) \sigma_i^z - \sin A(t) \sigma_i^y \right].
$$

(S.5)

When we consider the tunneling phenomena, it is useful to consider the response to the electric field $E(t)$ instead of the vector potential. The coupling with the electric field is introduced in the TI model as

$$
- \frac{E(t)}{2} \sum_i \sigma_i^z
$$

(S.6)

where $(1/2) \sum_i \sigma_i^z$ is identified as the electric dipole moment inside of dimer units.

The electric current operator is identified as

$$
\tilde{j}(t) = -\frac{1}{N} \frac{\delta \mathcal{H}}{\delta A(t)} = -t_0 \frac{1}{N} \sum_i \left[ \cos A(t) \sigma_i^x + \sin A(t) \sigma_i^y \right],
$$

(S.7)

where Eq. (S.5) is adopted as the coupling term with light. This vector-potential picture represented by the wavefunction $|\psi(t)\rangle$ is changed into the electric-field picture represented by $|\psi(t)\rangle$ where Eq. (S.6) is adopted. By introducing the unitary transformation $|\tilde{\psi}(t)\rangle = U(t)|\psi(t)\rangle$ with $U(t) = \exp[-iA(t) \sum_i \sigma_i^z / 2]$, the current operator in the electric-field picture is given by

$$
\tilde{j}' = -t_0 \frac{1}{N} \sum_i \sigma_i^y,
$$

(S.8)

which satisfies the relation $\langle \tilde{j}'(t) | \tilde{j}'(\tilde{t}) \rangle = \langle \psi(t) | \tilde{j} | \psi(t) \rangle$. 

Supplemental Material for “High-Harmonic Generation in a Correlated Electron System”

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interaction parameter dependence

In Fig. S.1 the Fourier transforms of the currents \( \langle \dot{j} \rangle_\omega \) in the case of \( V'/4\hbar_0 = 1.6 \) are presented. The infinite time-evolving block decimation (iTEBD) method with \( \chi = 200 \) is utilized to analyze the TI mode in the continuous-wave (cw) field. The threshold behavior of the HHG spectra with respect to amplitude of the vector potential, \( A_0 \), are commonly observed, and the threshold increases with increasing \( V' \).

finite size effects

In order to confirm the calculated results by the iTEBD method shown in MT, we show the results obtained by the exact diagonalization (ED) method based on the Lanczos algorithm. A periodic boundary condition is imposed on an \( N \)-site cluster. We introduce the cw light. A time profile of the induced current \( \langle \dot{j} \rangle_t \) and an intensity plot of the Fourier transform of current \( \langle \dot{j} \rangle_\omega \) calculated by the ED method are shown in Figs. S.2(a) and S.2(b), respectively. Overall features of \( \langle \dot{j} \rangle_t \) and \( \langle \dot{j} \rangle_\omega \)

almost reproduce the results shown in Figs. 1(d) and 2(a) in MT. We confirm the following two characteristics in \( \langle \dot{j} \rangle_\omega \): the HHG spectra show a plateau mainly between the upper and lower edges of the kink-antikink excitation spectra, i.e., \( 50 \leq \omega/\omega_0 \leq 130 \), and there is a threshold \( A_0 \sim 4 \) with respect to amplitude of light.

Results for the one-cycle pulse calculated by the ED method are shown in Fig. S.3 corresponding to Fig. 2(c) in MT calculated by the iTEBD method. The absorbed energy after the pulse irradiation \( \Delta E \) is plotted as a function of the electric field \( E_{\text{peak}} \) in Fig. S.3(a). We define peak height of the electric field \( E_{\text{peak}} = 0.928E_0 \) with \( E_0 = A_0(\omega_0 \omega_0) \). The exponential-like behavior mentioned in MT, i.e., \( \Delta E \propto \exp(-E_{\text{th}}/E_0) \) with the threshold electric field \( E_{\text{th}} \), is shown. Calculated data exceed \( E_{\text{peak}} \leq 0.1 \) are attributed to the numerical artifact. Weak multiple dip-and-hump structures appear in the \( \Delta E-E_{\text{peak}} \) curve. The energy level diagram under the static electric field \( E_s \) is shown in Fig. S.3(b), which is a part of Fig. 3(a) in MT. Each dip-and-hump structure corresponds to an anticrossing point between \( E_\beta \), which is adiabatically connected to the ground state at \( E_s = 0 \) and an excited energy level, as indicated by thin vertical lines. This implies that these structures originate from Landau-Zener tunneling processes. The absorbed energies calculated by the ED method are plotted as function of \( 1/E_0 \) in Fig. S.3(c) for \( N = 16 \) and 18. The dip-and-hump structure is weakened with increasing \( N \), and the data by the ED method tends to converge to that by the iTEBD method.
Calculated data sets almost reproduce the results obtained by the iTEBD method where the truncation parameter is chosen to be $\chi = 200$.

**OPTICAL ABSORPTION SPECTRA**

The optical absorption spectra are calculated by the iTEBD method. The energy increment after the weak irradiation ($A_0 = 0.0001$) is calculated in the real time evolution. The intensity map of the spectra is shown in Fig. S.4, which reproduces the previously known results schematically shown in Fig. 1(b) in MT, except for the region near $V'/4t_0 = 1$ where the data with enough accuracy are not obtained in the iTEBD method.

**HHG IN DIMER-MOTT PHASE**

In order to compare the HHG spectra in the polar CO state shown in MT, we show here the results of HHG in the dimer-Mott (DM) state. Intensity maps of the Fourier transforms of the current $\langle \hat{j} \rangle_\omega$ are calculated in the DM state. Results in the TI model and the spinless-fermion model are shown in Figs. S.5(a) and S.5(b), respectively. The current in $\omega/\omega_0 \approx 10$ and around $20 \lesssim \omega/\omega_0 \lesssim 70$ originates from the exciton excitation and the individual excitation of fermions, respectively. Owing to the space inversion symmetry, the HHG appears at $\omega = n\omega_0$ where $n$ is odd numbers. In the TI model, the HHG signal due to the exciton only appears, since the electrons are localized inside of the dimer units.

[1] M. Tsuchiizu (private communication).