High-harmonic generation in the Rice-Mele model: Role of intraband current originating from interband transition

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We consider high-harmonic generation (HHG) in the Rice-Mele model to study the role of the intraband current originating from the change of the intraband dipole via interband transition. This contribution, which has been often neglected in previous works, is necessary for the consistent theoretical formulation of the light-matter coupling. We demonstrate that the contribution becomes crucial when the gap is smaller than or comparable to the excitation frequency and the system is close to the half filling.

KEYWORDS: High-harmonic generation

1. Introduction

High-harmonic generation is a fundamental nonlinear optical phenomenon originating from strong light-matter coupling. It was originally observed and studied in atomic and molecular gases [1, 2]. Recently, HHG was observed in solids, such as semiconductors and semimetals, which extended the scope of the HHG research to solids [3–6]. One intriguing aspect of solids is that their properties can be controlled using active parameters such as temperatures, doping and pressure [7–11]. In order to correctly predict the dependence of HHG on these parameters, the consistent theoretical treatment of the light-matter coupling is necessary. A major approach to study HHG in solids is the semiconductor Bloch equations (SBEs) focusing on the several bands around the Fermi level [12–14]. However, the expression of SBEs depends on the gauges for the light and bases for electron states, which may lead to the inconsistency among the results obtained in terms of distinct choices. Recently, the relation between the different representations has been investigated in detail [15–17]. There is a term in the interband current that represents the change of the intraband dipole via interband transition although it is often neglected in the HHG analysis based on the well-used SBEs. Then a question arises; in which conditions this contribution is crucial for HHG? To answer this question, we numerically study HHG in the one-dimensional Rice-Mele model [18]. We discuss how important the often-neglected term is in the system when the gap-size and doping level is systematically changed.

2. Model and Method

We start with the one-dimensional Rice-Mele model [18] in the length gauge, whose Hamiltonian is

\[
\hat{H}_0 = \sum_i \left( \frac{Q_x - Q_y (-1)^i}{2} (c_i^\dagger c_{i+1} + \text{h.c.}) + \sum_i Q_{on} (-1)^i c_i^\dagger c_i - qE(t) \sum_i r_i c_i^\dagger c_i \right),
\]

where \( c_i^\dagger \) creates an electron at the \( i \)th site, \( Q_x \) is the averaged hopping between the nearest neighboring sites, \( Q_y \) is the hopping alternation, and \( Q_{on} \) is the staggered onsite energy. \( q \) is the charge,
2.1 Representation I: Dipole gauge expressed with the localized Wannier basis

In the dipole gauge, the light-matter coupling is taken into account through the Peierls phase as

$$\hat{H}^D(t) = \sum_k \left( \frac{Q_x - Q_y(-1)^{\frac{x}{2}}}{2} \left( e^{-i q A_I(t)} c^\dagger_i c_{i+1} + \text{h.c.} \right) + Q_{\text{a}}(-1)^{\frac{x}{2}} c^\dagger_i c_i \right),$$

(2)

where $a$ is the bond length, and $A(t)$ is the vector potential. Namely, $E(t) = -\partial_t A(t)$. Using the Fourier transformations, the creation operators in the $\alpha (= A, B)$ sublattice are given by $c^\dagger_{\alpha k} = \frac{1}{\sqrt{N}} \sum_{i \in \alpha} e^{i k \cdot r_i} c^\dagger_i$. The Hamiltonian in the $k$-space is given as

$$\hat{H}^D(t) = \sum_k \left( c^\dagger_{\alpha k} c^\dagger_{\beta k} \right) h(k - q A(t)) \left( c_{\alpha k} c_{\beta k} \right),$$

(3)

$$h(k) = Q_x \cos \left( \frac{k}{2} \right) \sigma_x + Q_y \sin \left( \frac{k}{2} \right) \sigma_y + Q_{\text{relax}} \sigma_z,$$

(4)

where we set $a = \frac{1}{2}$ and $\sigma_m (m = x, y, z)$ are the Pauli matrices. In the SBE approach, we focus on the single-particle density matrix (SPDM) $\rho^D_{\alpha \beta k}(t) = \langle c^\dagger_{\beta k}(t) c_{\alpha k}(t) \rangle$, where $\langle \cdots \rangle$ is the expectation value with the grand canonical ensemble and $c^\dagger(t)$ indicates the Heisenberg representation of $c^\dagger$. The von Neumann equation of SPDM (or simply SBE) is expressed as

$$\partial_t \rho^D_{\alpha \beta k} = -i [h(k(t)), \rho^D_{\alpha \beta k}(t)] + \partial_t \rho^D_{\alpha \beta k}^{\text{relax}},$$

(5)

where $\rho^D_{\alpha \beta k}(t)$ is the matrix with elements $\rho^D_{\alpha \beta k}(t), k(t) = k - q A(t)$ and $\hat{h}$ is set unity. The last term represents relaxing and dephasing processes originating from electron-electron interactions, electron-phonon interactions and scattering with impurities. The microscopic evaluation of $\partial_t \rho^D_{\alpha \beta k}^{\text{relax}}$ is computationally expensive. Instead, in this paper, we set $\partial_t \rho^D_{\alpha \beta k}^{\text{relax}} = -\partial_t \rho^D_{\alpha \beta k}^{\text{relax}} = \rho^D_{\alpha \beta k}(t) - \rho^D_{\alpha \beta k}(t) = \sum_{\omega} \rho^D_{\alpha \beta k}^{\text{relax}}(\omega) / T_1$ and take account of the relaxation and dephasing processes phenomenologically. Here $\rho^D_{\alpha \beta k}^{\text{relax}}(t)$ represents the SPDM in the equilibrium state. In this representation, the operator of the current is expressed as

$$\hat{J}(t) = \sum_k \psi^\dagger_k \left[ \partial_t h(k(t)) \right] \psi_k,$$

(6)

where $\psi_k^\dagger = [c^\dagger_{\alpha k}, c^\dagger_{\beta k}]$. Note that the expectation value of the current can be obtained by the SPDM. The intensity of HHG is also evaluated from the current $J(t)$ as $I_{\text{HHG}}(\omega) = |\omega J(\omega)|^2$, where $J(\omega)$ is the Fourier component of $J(t)$. Since the expression of $h(k)$ is easily evaluated, this representation is beneficial for the numerical simulation of the time evolution of the system excited by the light. However, to classify distinct contributions to HHG, it is more convenient to consider the basis set that diagonalizes $h(k(t))$ as the following representations.
2.2 Representation II: Dipole gauge expressed with the Houston basis

We introduce the unitary matrix $U(k)$ that satisfies $U(k)\hat{h}(k)U(k) = \epsilon(k)$, where $\epsilon(k) = \text{diag}[\epsilon_c(k), \epsilon_v(k)]$. The subscripts $c$ and $v$ refer to the conduction band and the valence band, respectively. Considering the time dependent unitary transformation as $\hat{\psi}_k \rightarrow U(k(t))\hat{\psi}_k$, we obtain the Hamiltonian

$$\hat{H}^H(t) = \sum_k \hat{\psi}_k^\dagger(t) \epsilon(k(t)) \hat{\psi}_k - qE(t) \sum_k \hat{\psi}_k^\dagger d(k(t)) \hat{\psi}_k,$$

where $\hat{\psi}_k = [\hat{b}_k^c, \hat{b}_k^v]$. $d(k) = iU(k)\hat{h}_kU(k)$ is the Berry connection, which plays the role of the dipole matrix. In this representation, the von Neumann equation for $\rho_{mn,k}^H(t) = \langle \hat{b}_{kn}(t)\hat{b}_{km}(t) \rangle$ becomes

$$\partial_t \rho_{kn,k}^H(t) = -i[\epsilon(k(t)) - qE(t)d(k(t)), \rho_{kn,k}^H(t)].$$

Actually, this representation is essentially equivalent to the representation III, which will be shown below.

2.3 Representation III: Length gauge expressed with the band basis

Now we express the Hamiltonian (3) in the length gauge, using the band basis $[c_{kc}^\dagger, c_{kv}^\dagger] = [c_{kA}^\dagger, c_{kB}^\dagger]U(k)$. $\hat{H}^L(t)$ is given as

$$\hat{H}^L(t) = \sum_k (c_{kc}^\dagger c_{kv}^\dagger) \epsilon(k) \left(\begin{array}{c} c_{kc} \\ c_{kv} \end{array}\right) - E(t) \cdot \hat{P},$$

where $\hat{P}$ is the polarization operator. This operator can be divided into the intra- and interband polarizations as $\hat{P} = \hat{P}_{\text{ra}} + \hat{P}_{\text{er}}$, where

$$\hat{P}_{\text{ra}} = q \sum_k \sum_n d_{mn}(k)c_{kn}^\dagger c_{km},$$

$$\hat{P}_{\text{er}} = q \sum_k \sum_{n \neq m} d_{mn}(k)c_{kn}^\dagger c_{km},$$

where $d(k) = iU(k)\hat{h}_kU(k)$. The current is expressed as the change of the polarization as $\hat{J}(t) = -i[\hat{P}, \hat{H}^L(t)]$. Thus, the intraband current is defined as $\hat{J}_{\text{ra}}(t) = -i[\hat{P}_{\text{ra}}, \hat{H}^L(t)]$ and the interband current is defined as $\hat{J}_{\text{er}}(t) = -i[\hat{P}_{\text{er}}, \hat{H}^L(t)]$. The intraband current $\hat{J}_{\text{ra}}(t) = \hat{J}_{\text{ra}}^{(I)}(t) + \hat{J}_{\text{ra}}^{(II)}(t)$ is given by

$$\hat{J}_{\text{ra}}^{(I)}(t) = q \sum_k \sum_n \partial_k \epsilon(k) c_{kn}^\dagger c_{km},$$

$$\hat{J}_{\text{ra}}^{(II)}(t) = -qE(t) \sum_k \sum_{n \neq m} (\partial_k [d_{mn}(k)] + i(d_{mn}(k) - d_{mm}(k))^\dagger c_{kn}^\dagger c_{km}.$$

We find that $\hat{J}_{\text{ra}}^{(I)}(t)$ consists of the diagonal components of $c_n^\dagger c_m$, while $\hat{J}_{\text{ra}}^{(II)}(t)$ consists of the off-diagonal components. The current $\hat{J}_{\text{ra}}^{(I)}(t)$ originates from $-i[\hat{P}_{\text{ra}}, -E(t) \cdot \hat{P}_{\text{er}}]$, and represents the change of the intraband dipole via interband transition. The interband current $\hat{J}_{\text{er}}(t)$ is given as

$$\hat{J}_{\text{er}}(t) = -iq \sum_k (c_{kc}^\dagger c_{kv}^\dagger) [d(k), \epsilon(k)](c_{kc}^\dagger c_{kv}) + qE(t) \sum_{n \neq m} \sum_k (\partial_k [d_{mn}(k)] - i(d_{mn}(k) - d_{mm}(k))^\dagger c_{kn}^\dagger c_{km}. $$

The von Neumann equation for $\rho_{mn,k}^{LB}(t) = \langle c_{kn}(t)c_{km}(t) \rangle$ in this representation is

$$\partial_t \rho_{kn,k}^{LB}(t) = -i[H^{LB}(k,t), \rho_{kn,k}^{LB}(t)] - (E(t) \cdot \nabla_k) \rho_{kn,k}^{LB}(t).$$
where \( \hat{J}_{\text{LB}}^I(k,t) = \epsilon(k) - E(t) d(k) \). Introducing \( \hat{\rho}^I_{k}(t) \equiv \rho^I_{k-qA(t)}(t) \), we have

\[
\partial_t \hat{\rho}^I_k (t) = -i [\hat{h}^I_k(k-qA(t)), \hat{\rho}^I_k(t)].
\]

This equation is the same as Eq. (8) for SPDM in the representation II. Since the initial condition of SPDM is also the same for the representations II and III, we have \( \hat{\rho}^I_k(t) = \hat{\rho}^I_{k-LB}(t) \).

The SBE in the form of Eq. (15) has often been used up to now, where the intraband and interband currents are evaluated separately. However, in this treatment, some terms of the current have been often overlooked [15, 17], e.g., the current contribution originating from the change of the intraband dipole via the interband transition \( \hat{J}_{\text{ra}}^{(\text{II})}(t) \). In the following, we examine the contributions of different types of currents, performing the simulation based on the representation I.

3. Results

![Fig. 1.](image)

(a) Band structure of the Rice-Mele model with \( Q_y = 3 \), and \( Q_x = Q_{on} = 0.25 \). \( E_{gap} \) indicates the minimum band gap. (b) \( \alpha_n \) as a function of \( n_{\text{odd}} \) and \( E_{gap}/\Omega \). (c) \( \alpha_n \) as a function of \( n_{\text{even}} \) and \( E_{gap}/\Omega \). For (b) and (c), we set \( Q_y = 3 \) and the chemical potential \( \mu = 0 \) (the half filling), and keep \( Q_x = Q_{on} \). The value of \( E_{gap} \) reflects the change of the parameters \( Q_x \) and \( Q_{on} \). The parameters of the electric field are \( \Omega = 0.2 \), \( A_0 = 1 \), \( \sigma = 300 \), \( t_0 = 1200 \) and \( T_1 = 4\pi \).

To discuss how important the well-neglected current \( \hat{J}_{\text{ra}}^{(\text{II})}(t) \) is in the HHG analysis, we examine the time-evolution of the system after the electric field pulse is introduced. Here, we consider the electric field pulse in the Gaussian form as \( A(t) = A_0 \exp(-\frac{(t-t_0)^2}{2\sigma^2}) \sin(\Omega(t-t_0)) \). To reveal the role of \( \hat{J}_{\text{ra}}^{(\text{II})}(t) \) in HHG, we introduce \( \alpha_n \) as

\[
\alpha_n = \frac{\int_{-\delta\pi}^{\delta\pi} d\omega \, J(\omega)}{\int_{-\delta\pi}^{\delta\pi} d\omega \, J_{\text{simp}}(\omega)},
\]

where \( \hat{J}_{\text{simp}}(t) = \hat{J}_{\text{ra}}^{(\text{I})}(t) + \hat{J}_{\text{er}}(t) \) is a part of the full current operator and \( J_{\text{simp}}(\omega) \) is the Fourier component of its expectation value. In practice, we set \( \delta = 0.5 \). The contribution from \( \hat{J}_{\text{ra}}^{(\text{II})}(t) \) is crucial when \( \alpha_n \) is far away from unity. First, we focus on the half-filled system (\( \mu = 0 \)) and study the band gap dependence under the condition \( Q_x = Q_{on} \). In the case \( Q_y = 3 \) and \( Q_x = Q_{on} = 0.25 \), its band structure is shown in Fig. 1(a), where the band gap \( E_{gap} \sim 0.7 \). In the system, when both \( Q_x \) and \( Q_{on} \) are small, the system approaches the system with the inversion symmetry, and thereby the intensity of odd harmonics is larger than even one. In Figs. 1(b) and 1(c), we show the result for
\( \alpha_n \) with odd and even \( n \), respectively. We find that \( \alpha_n \) becomes much smaller than unity when \( E_{\text{gap}} \) is smaller than or comparable to the excitation frequency. On the other hand, when \( E_{\text{gap}} \) is large, \( \alpha_n \) is close to unity. These results imply that, when the system is half filled and the band gap is smaller than or comparable to the excitation frequency, the cancelation between the intraband and interband currents is severe and the careful treatment of \( \hat{J}_{ra}(t) \) is needed to evaluate the HHG spectrum correctly.

\[
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\]

Next, we discuss the doping dependence of \( J_{ra}^{(1)}(t) \), considering the system with \( Q_y = 3 \) and \( Q_n = Q_x = 1 \times 10^{-4} \), where a tiny gap appears in the single-particle spectrum, as shown in Fig. 2(a). In the case, the system is almost symmetric under the inversion operation. Therefore, we focus on relevant odd harmonics. Figures 2(b) and 2(c) show \( \alpha_n \) as a function of the chemical potential for odd \( n \). We find that, as for the first harmonics, \( \alpha_1 \) is always close to unity, implying that \( J_{ra}^{(1)}(t) \) is irrelevant. On the other hand, different behavior appears in the higher harmonics (\( n = 3, 5 \)). When the system is half filling (\( \mu = 0 \)), \( \alpha_n \) is far away from unity, as discussed above. When the holes are doped in the system, \( \alpha_n \) slightly increases. When \( \mu \approx \mu_c \), \( \alpha_n \) (\( n = 3, 5 \)) increases suddenly, and it reaches unity, as shown in Figs. 2(b) and 2(c). Here, we introduce \( \mu_c \) as \( \epsilon_v(k = \pm A_0) \). These results suggest that the contribution of \( J_{ra}^{(1)}(t) \) is crucial when the system is close to the half filling and the band gap is small enough to the excitation frequency.

Now we study the distinct contributions for the currents in detail. To this end, we introduce

\[
\beta_n = \frac{\int \omega J_{er}(\omega) d\omega}{\int \omega J_{ra}(\omega) d\omega}, \quad \gamma_n = \frac{\int \omega J_{ra}^{(1)}(\omega) d\omega}{\int \omega J_{ra}(\omega) d\omega}, \quad (18)
\]

which allows us to discuss the role of the currents \( \hat{J}_{er}(t) \) and \( \hat{J}_{ra}^{(1)}(t) \) in the HHG spectrum. We show these quantities as a function of the chemical potential in Figs. 2(d) and 2(e). It is found that \( \beta_n \) and

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**Fig. 2.** (a) Band structure of the model with \( Q_y = 3 \) and \( Q_n = Q_x = 1 \times 10^{-4} \) for which we study the doping dependence of HHG. (b,c) The corresponding \( \alpha_n \) as a function of chemical potential. (d) The corresponding \( \beta_n \) as a function of chemical potential. (e) The corresponding \( \gamma_n \) as a function of chemical potential. In all cases, the parameters of the electric field are \( \Omega = 0.2, \sigma = 300, t_0 = 1200 \) and \( T_1 = 4\pi \).
\( \gamma_n \) are qualitatively similar to each other. Namely, when \( \mu \gtrsim \mu_c \), they are almost constant, while they decrease abruptly around \( \mu = \mu_c \). The results imply that the contribution of \( \tilde{J}^{(I)}_{\text{ra}}(t) \) is dominant for \( \mu \lesssim \mu_c \), and both \( \tilde{J}^{(II)}_{\text{ra}}(t) \) and \( \tilde{J}_{\text{er}}(t) \) are dominant for \( \mu \gtrsim \mu_c \). This change of the dominant contribution in HHG around \( \mu_c (= \epsilon_s (k = \pm A_0)) \) can be understood as follows. In the present system, one can expect that the interband transition is strongly suppressed when \( \mu \lesssim \mu_c \). This is because no electrons can reach the Gamma point, where the band gap is minimum, during the pulse. Note that when an electron is excited by the electric field, its momentum is shifted by the vector potential. In addition, \( \tilde{J}^{(II)}_{\text{ra}}(t) \) corresponds to the change of the intraband dipole via interband transition and \( \tilde{J}_{\text{er}}(t) \) corresponds to the change of the interband dipole. Therefore, the suppression of the interband transition implies that these contributions become less important.

4. Summary

To summarize, we have studied the effects of the often-neglected current \( \tilde{J}^{(II)}_{\text{ra}}(t) \), which originates from the change of the intraband dipole via interband transition, on HHG in the one-dimensional Rice-Mele model. When the system is close to the half filling and the band gap is smaller than or comparable to the excitation frequency, the contribution becomes crucial. Our results suggest the importance of the full evaluation of the currents when one studies HHG in small gap systems such as graphene, Weyl semimetals and metallic carbon nanotubes.

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