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Detuning dependence of higher-order harmonic generation in monolayer transition metal dichalcogenides

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Abstract

We theoretically investigate detuning-dependent properties of high harmonic generation (HHG) in monolayer transition metal dichalcogenides (TMDCs). In contrast to HHG in conventional materials, TMDCs show the both perpendicular and parallel emissions with respect to the incident electric field. We also find that such an anomalous emission can be controlled by the frequency detuning of incident fields. This peculiar phenomenon enables us to realize novel optical devices.

1. Introduction

Atomically thin two-dimensional materials have been widely investigated in recent years because of their potential utilities for future electronics applications. The transition metal dichalcogenides (TMDCs) are one of the most typical materials, showing the two-dimensional hexagonal lattice with M (M = Mo or W) and X (X = S or Se) atoms and with the space inversion breaking. The space inversion symmetry breaking is the origin of the energy band-gap appearing at the $K^{\pm}$ points and provides a novel platform to explore unique electric and optical properties of TMDCs [1-6], including a valley-dependent selection rule for interband excitations of Bloch electrons with circularly polarized electric fields. Actually, some recent experiments [7-11] have shown the peculiar characteristics of high harmonic generation (HHG) in TMDCs: HHG could be identified not only in the parallel but also in the perpendicular directions with respect to the incident electric fields (Fig. 1). Moreover, they are quite sensitive to the orientation of lattice structures and show the reversed-phase dependence on the crystallographic orientation. Thus, unique characteristics of TMDCs are expected to have the potential utilities for optical technologies and establishment of the control method for HHG would be demanded urgently for opening new optical applications of TMDCs.

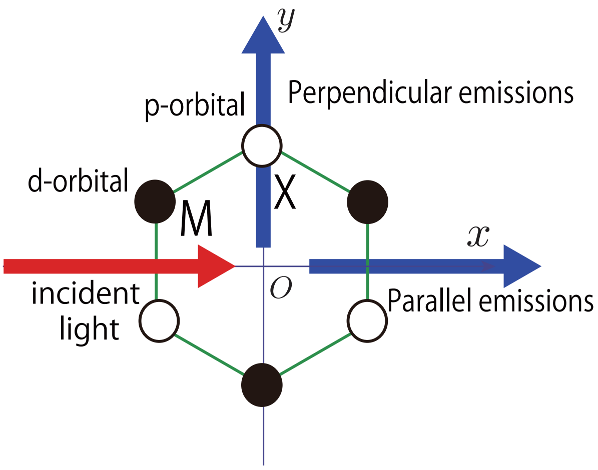

Fig.1 Schematic diagram of HHG in TMDCs.

In this presentation, we will theoretically propose a way to artificially control both parallel and perpendicular emissions of HHG in TMDCs, by tuning the frequency of the incident electric fields. We show the *n*th ordered harmonics to be enhanced when the frequency of the incident electric fields is set to one *n*th. This peculiar phenomenon would provide a way to control the both perpendicular and parallel emissions of HHG.

2. Theory

The theoretical framework utilized in this presentation was constructed by applying the theory we have established previously [12-14] to the case of TMDCs. The framework is based on the tight-binding model, where the honeycomb lattice MX_2 is constructed with M (S, Se, Te) and X (Mo, W) atoms. In this model, we will assume the wave-functions of M and X atoms as $\phi_A(x) = p_x(x) + ip_y(x)$ and $\phi_B(x) = d_{xy}(x) + i\tau^z d_{x^2-y^2}(x)$, respectively. Here, τ^z is the variable describing the state at the $K^{\pm}$ points. In addition, we will define the difference of the onsite energy for each atom as m. The Rabi frequency in this model can be represented as $\Omega_{\mathrm{R}}(\boldsymbol{k},t) = (e/m_0 c)\Sigma_i e^{i\boldsymbol{k}\cdot\boldsymbol{\delta}_i} \int d^2x\, \phi_B^*(\boldsymbol{x})\boldsymbol{A}(t)\cdot\boldsymbol{p}\phi_A(\boldsymbol{x}-\boldsymbol{\delta}_i)$. Here, $\boldsymbol{\delta}_i$ and $\boldsymbol{A}(t)$ are the lattice vector and vector potential, respectively. Considering the above expressions and performing the same procedure as in Refs [12-14], we can derive the the time evolution equations of the carrier densities $f_{\boldsymbol{k}}^{\sigma} = \langle\sigma_{\boldsymbol{k}}^{\dagger}\sigma_{\boldsymbol{k}}\rangle(\sigma = e,h)$ and the polarization $P_{\boldsymbol{k}} = \langle h_{-\boldsymbol{k}}^{\dagger}e_{\boldsymbol{k}}\rangle$ with the Bloch wave-vector $\boldsymbol{k}$. The numerical solutions of these equations give the time evolutions of the distributions of the carrier densities $f_{\boldsymbol{k}}^{\sigma} = \langle\sigma_{\boldsymbol{k}}^{\dagger}\sigma_{\boldsymbol{k}}\rangle$ and polarization $P_{\boldsymbol{k}} = \langle h_{-\boldsymbol{k}}^{\dagger}e_{\boldsymbol{k}}\rangle$ in two-dimensional $\boldsymbol{k}$ space. Utilizing these distributions, the time evolution of the generated currents along the *x*- and *y*- axes can be calculated using $J_\nu(t) = -c\langle\partial H_I/\partial A_\nu\rangle(\nu = x, y)$. Thus, we can obtain the time evolutions of the current in the form, $J(t) = \left(J_x(t), J_y(t)\right)$, and can calculate HHG spectra from $I(\omega) = \omega^2|J(\omega)|^2$, where $J(\omega) = \left(J_x(\omega), J_y(\omega)\right)$ is the Fourier transform of $J(t)$.

3. Results and Discussions

Our numerical calculation shows that the both parallel and perpendicular emissions exist with respect to the incident electric fields. The parallel emissions of HHG mainly include only odd-ordered harmonics while the perpendicular emissions include the even-ordered ones. These tendencies can be attributed to the unique wave number dependence of the Rabi frequency, which is fundamentally determined by the lattice structures and wave-functions of each atoms. [14]. We plotted in Fig. 2 the detuning dependence of the parallel emissions of HHG for the third- (blue dots), fifth- (orange dots), and seventh-ordered (green dots) harmonics. In this numerical calculation, we fix the Rabi frequency as $\Omega_{R0} = 0.1\omega_0$ and vary the ratio of band-gap energy and the frequency of the incident electric field, i.e., $E_g/\hbar\omega_0$. This figure clearly indicates that the third, fifth, and seventh-ordered harmonics have maximum peaks at $E_g/\hbar\omega_0 = 3, 5,$ and 7, respectively. We also plotted in Fig. 3 the detuning dependence of the perpendicular emissions of HHG for the second- (blue dots), fourth- (orange dots), and sixth-ordered (green dots) harmonics. Similar to the parallel emissions, the second, fourth, and sixth-ordered harmonics have maximum peaks at $E_g/\hbar\omega_0 = 2, 4,$ and 6. These results indicate that the HHG can be maximized at the resonant frequencies and the emission amplitudes can be dramatically controlled by the detuning . This peculiar phenomenon would provide a way to control HHG in TMDCs.

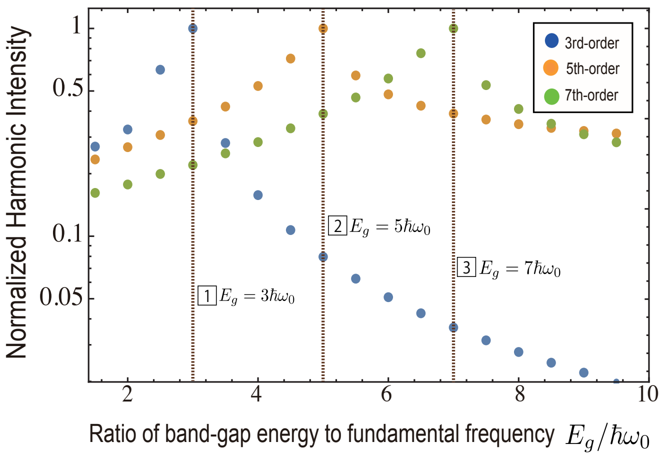

Fig.2 Detuning-dependent properties of third, fifth, and seventh-harmonics in the parallel emissions with respect to the incident electric fields.

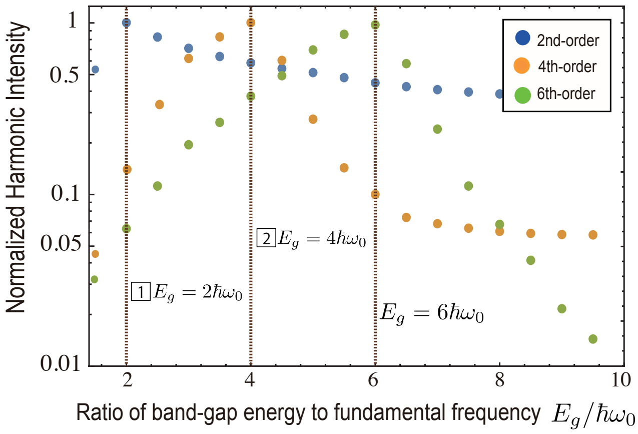

Fig.3 Detuning-dependent properties of second, forth, and sixth harmonics HHG in the perpendicular emissions with respect to the incident electric fields.

4. Conclusions

We theoretically investigate the detuning dependence of HHG in TMDCs. We found that the both parallel and perpendicular emissions show the drastic enhancement at the resonant frequency. In addition, the emission amplitudes can be effectively controlled by the frequency detuning of incident fields. Our peculiar results would provide new functional optical devices based on TMDCs, such as sensors, frequency modulators and frequency converters in optical and THz regimes.

Acknowledgements

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