



Extended high-harmonic spectra through a cascade resonance in confined quantum systems

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The study of high-harmonic generation in confined quantum systems is vital to establishing a complete physical picture of harmonic generation from atoms and molecules to bulk solids. Based on a multilevel approach, we demonstrate how intraband resonances significantly influence the harmonic spectra via charge pumping to the higher subbands and thus redefine the cutoff laws. As a proof of principle, we consider the interaction of graphene nanoribbons, with zigzag as well as armchair terminations, and resonant fields polarized along the cross-ribbon direction. Here, this effect is particularly prominent due to many nearly equiseparated energy levels. In such a scenario, a cascade resonance effect can take place in high-harmonic generation when the field strength is above a critical threshold, which is completely different from the harmonic generation mechanism of atoms, molecules, and bulk solids. We further discuss the implications not only for other systems in a nanoribbon geometry, but also systems where only a few subbands (energy levels) meet this frequency-matching condition by considering a generalized multilevel Hamiltonian. Our study highlights that cascade resonance has a fundamentally distinct influence on the laws of harmonic generation, specifically the cutoff laws based on laser duration, field strength, and wavelength, thus unraveling additional insights in solid-state high-harmonic generation.

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I. INTRODUCTION

High-harmonic generation (HHG), originally studied in the gas phase for atoms and molecules [1–3], has led to the creation of isolated attosecond pulse [4–7], thus vigorously promoting the development of ultrafast technology [8–10]. Recently, the study of HHG has been extended to condensed-matter systems [11–38]. Since the first experimental observation in 2011 [11], solid-state HHG has gained immense interest due to potential applications, such as in compact ultrafast light sources [39,40]. In addition, it may serve as a tool to probe microscopic properties of matter, such as band structures [37,41,42], valence electron potentials [43], Berry curvatures [44,45], and phase transitions [46–49].

Depending on the intensity and frequency of the applied electric field, together with details of the band structure, even nonperturbative mechanisms can be responsible for solid-state HHG [50]. In this case, solid-HHG is believed to have

contributions from dynamical intraband and interband processes involving a k -space motion of Bloch electrons, typically described by a three-step model [12,13]. Nevertheless, a clear understanding of the underlying mechanism(s) for solid-state HHG is still under debate [14,50,51].

For a confined quantum system, due to a lack of translational symmetry, the spectrum consists of only a discrete set of energy levels instead of bands. It is known that the HHG spectra in quantum dots can be influenced by confinement conditions such as size and/or coupling parameters in coupled quantum dots [52,53]. One of the distinct advantages of confined quantum systems is the tunability of its energy spectra and wave functions by tailoring its size [54,55], or by external parameters, such as gate voltage or magnetic field [56–58].

A rather interesting situation is realized in partially confined systems, such as quasi-one-dimensional (quasi-1D) systems, which are confined in one dimension but periodic in others, resulting in a series of subbands in the band structure [59]. Therefore, such systems possess properties of both bulk and finite systems. The most notable example is perhaps the graphene nanoribbon (GNR) [60–65]. Depending on the polarization of the electric field, different aspects of HHG, corresponding to bulk and/or confined quantum systems, can be explored. Arguably, the phenomenology of HHG in such a confined quantum system, therefore, forms a bridge between atoms, molecules, and bulk solids. A detailed understanding of the underlying mechanism in such quantum systems may

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not only shed light on the HHG phenomenology in bulk solids, but it may also unravel new phenomenology and HHG response in confined systems.

Understanding the HHG phenomenology in such systems necessitates going beyond the two-level system (equivalent to the commonly employed two-band studies of the bulk solid-state HHG), thereby raising a number of conceptual questions: For a laser field polarized along the confining direction, what would happen if the laser frequency ω_0 approximately matches the subband gap, i.e., $\omega_0 \approx \Delta_{\text{sg}}$? Does it cause a resonance resembling Rabi flopping in a two-level system [66,67]? If many such (approximately) equiseparated subbands are present, does the resonance involve multiple subbands? If so, could this resonant excitation affect the harmonic process remarkably? In addition, Hansen *et al.* indicate that the observed cutoff law on HHG transits from atomic to solid-state type with increasing system size in a model 1D chain [68]. Does this phenomenon hold even in the presence of resonance?

Here, by considering a multilevel model and solving the time-dependent Schrödinger equations, we demonstrate that subband resonance can lead to remarkable effects in the HHG spectra. We find that this effect is especially prominent in GNRs due to the presence of nearly equiseparated bands. To this end, as a proof of principle, we consider GNRs interacting with a resonant field polarized along the cross-ribbon (confined) direction where it leads to *cascade resonance*, a multilevel phenomenon involving almost all the valence and conduction subbands, and enhanced HHG spectra well beyond the current cutoff laws.

Specifically, the plateaus of the harmonic spectra are broadened significantly when the laser frequency matches the subband gap. The cascade resonance effect causes the excited electrons to gradually accumulate near the Dirac points also in higher-energy subbands (charge pumping), eventually leading to the extended HHG spectra. Subsequently, we establish the conditions for the occurrence of cascade resonance in GNRs. Our analysis indicates that the occurrence of cascade resonance requires a threshold field strength in addition to satisfying the frequency-matching condition. In addition, we formulate the dependence of a harmonic cutoff in GNRs on laser duration, field strength, and wavelength when the cascade resonance occurs. These dependencies are significantly different from those of HHG in atoms, molecules, and bulk solids currently studied. Finally, we discuss the possibilities for cascade resonance in other materials by considering a general multilevel model and implications for HHG spectra.

II. HHG IN GRAPHENE NANORIBBONS

GNR is a quasi-1D material extending in two directions—ribbon (x) and cross-ribbon (y) directions [60–65]. GNR with armchair edges (AGNR) on both sides is classified by the number of dimer lines (N_a) across the ribbon width, as shown in Fig. 1(a). The unit cell of AGNR consists of two chains, P and Q . Likewise, GNR with zigzag-shaped edges (ZGNR) on both sides is classified by the number of the zigzag chains (N_z) across the ribbon width [Fig. 1(b)]. The chain in the unit cell of ZGNR is labeled by C . We refer to GNR with N_a armchair dimer lines as N_a -AGNR, and GNR with $N_z/2$ zigzag chains

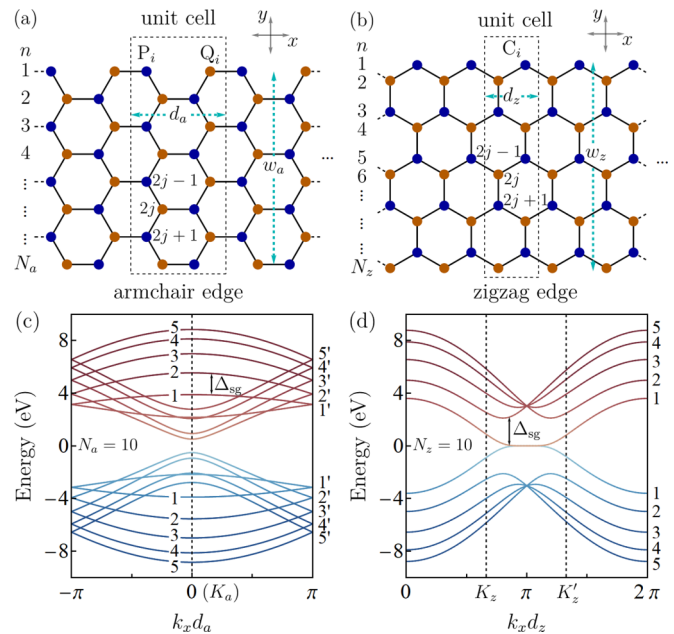


FIG. 1. (a),(b) Structure of graphene nanoribbons with (a) armchair edges and (b) zigzag edges. Blue and orange cycles represent the two nearest-neighbor carbon atoms. The boundary condition is periodic in the x direction but is open in the y direction. (c),(d) Band structure of (c) 10-AGNR and (d) 10-ZGNR. Red (blue) curves stand for the subbands belonging to the conduction (valence) band. Δ_{sg} denotes the subband gap. $K_a = 0$ and $K'_a = 2\pi/3d_a$, $4\pi/3d_a$ are Dirac points, respectively, for the armchair and zigzag GNRs if the boundary condition along the transverse (y) direction is periodic. w_a (w_z) is the width of the armchair (zigzag) nanoribbon. d_a (d_z) is the distance of the armchair (zigzag) unit cell.

as N_z -ZGNR. A tight-binding model of GNR is presented in Appendix A.

Since the ribbon is macroscopically large along the x (longitudinal) direction, continuous band structure can be obtained across the Brillouin zone (BZ), as shown in Figs. 1(c) and 1(d). By contrast, in the y (transverse) direction, transverse confinement gives rise to a discrete set of subbands, which is one of the typical features for electronic structure of nanoribbons. Following previous convention, we adopt $J(s)$ as the index notation for the ribbon subbands, see Figs. 1(c) and 1(d), where $J = 1, 2, \dots, N$ is the band number and s is the band type, with “ c ” and “ v ” representing conduction and valence bands, respectively. Different from subbands of ZGNR, the subband indices of the AGNR in Fig. 1(c) are classified into two groups labeled by $\{J\}$ and $\{J'\}$, respectively, where subbands $J(s)$ and $J'(s)$ merge at the boundaries of the BZ. Atomic units (a.u.) are used throughout the paper unless otherwise indicated.

Recently, there has been a growing interest in studying HHG from GNRs or similar nanoribbons [69–73] due to the diverse electronic properties of GNRs, which arise particularly from different edge geometries, viz., zigzag and armchair edges. When the laser field is polarized along the ribbon direction, the bulk aspects of GNRs are reflected in the harmonic spectra. For example, the edge states of ZGNR enhance the emitting efficiency of low-order harmonics [70]; the on-site

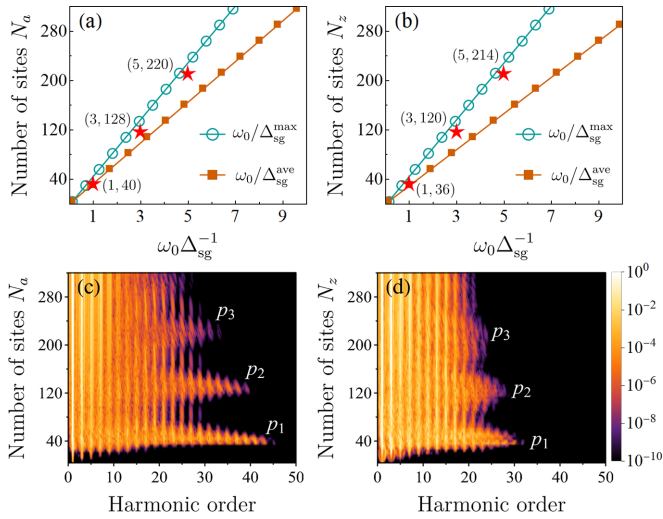


FIG. 2. (a),(b) Ratios of the driver frequency ω_0 and subband gaps Δ_{sg} at Dirac points as a function of system size N for (a) AGNRs and (b) ZGNRs. Lines with symbol \circ (\blacksquare) represent the variation of the maximum (average) value of subband gaps. (c),(d) Harmonic spectra vs number of sites N in (c) AGNRs and (d) ZGNRs, respectively, using a driver with $n_{\text{cyc}} = 16$, $\omega_0 = 0.0152$ a.u. ($\lambda = 3 \mu\text{m}$), and $E_0 = 0.0012$ a.u. ($I_0 = 5.04 \times 10^{10} \text{ W/cm}^2$). The color bar characterizes the intensity of harmonics. The resonance peaks (p_1 , p_2 , and p_3) in (c) and (d) are marked in (a) and (b), respectively, in the form of red stars.

potential, breaking mirror symmetry, causes the perpendicular harmonic emission [72,73]. It is easy to know that the cutoffs of longitudinal harmonic spectra scale linearly to the field strength and wavelength, as shown in Appendix D, which is similar to the HHG in bulk graphene. However, the finite-size effects [68,74] in GNR along the cross-ribbon direction have not yet been fully examined.

In GNRs, the HHG generated by a transverse field can arguably be more interesting than that generated by a longitudinal field. Quantum confinement effects reflect on HHG when the applied laser field is polarized along the cross-ribbon direction. Specifically, the nearly equal-energy spacing subbands play a role and induce a resonance excitation over the subbands. In general, however, optical transitions between two subbands are not always allowed. The optical selection rules for GNRs are a result of the wave-function parity factor $(-1)^J$, where J is the subband index that was mentioned before. A detailed derivation and discussion of the optical selection rules is outlined in Appendix B.

A. Size dependence of the transverse HHG spectra

The subbands near the Dirac points are approximately equidistant. We denote the gap between nearest-neighbor subbands as Δ_{sg} , shown in Figs. 1(c) and 1(d). It is characterized by $\Delta_{sg} \sim N^{-1}$. We can see in Figs. 2(a) and 2(b) that both $(\Delta_{sg}^{\max})^{-1}$ and $(\Delta_{sg}^{\text{ave}})^{-1}$ for AGNR and ZGNR are approximately linear with the number of sites N , where Δ_{sg}^{\max} is the maximum subband gap, and Δ_{sg}^{ave} is the average value of all subband gaps. Both Δ_{sg}^{\max} and Δ_{sg}^{ave} are presented since the subband gaps at the Dirac points are not precisely equal.

Figures 2(c) and 2(d) display the size dependence of harmonic spectra for AGNR and ZGNR, respectively, while the driver field is polarized along the transverse direction. The laser frequency and intensity are fixed at $\omega_0 = 0.41$ eV (0.0152 a.u.) and $I_0 = 5.04 \times 10^{10} \text{ W/cm}^2$, respectively. In the spectrum of AGNR-HHG as shown in Fig. 2(c), the harmonic cutoff is drastically extended for widths $N_a = 40, 128$, and 220 (ribbon width $w_a \approx 4.8, 15.6$, and 26.9 nm), forming three peaks p_1 , p_2 , and p_3 , respectively. These three peaks are depicted in Fig. 2(a) as red stars, whose abscissas correspond to $\Delta_{sg} = \omega_0, \omega_0/3$ and $\omega_0/5$. For the first peak, at $N_a = 40$, it is found in Fig. 2(a) that $\Delta_{sg}^{\text{ave}}(40) \lesssim \omega_0 \lesssim \Delta_{sg}^{\max}(40)$, i.e., the laser frequency matches the subband gap [$\omega_0 \approx \Delta_{sg}(40)$]. Therefore, the harmonic spectrum is affected significantly when the resonance condition ($\omega_0 = \Delta_{sg}$) is met. Two additional peaks at $N_a = 128$ and 220 reveal that the resonant excitation occurs not only when $\Delta_{sg}(40) \approx \omega_0$, but also when $\Delta_{sg}(N_a) \approx \omega_0/3$ and $\omega_0/5$. This can be understood by the optical selection rule of AGNR in Appendix B when subbands are from the same group. The intraband (interband) transitions are permitted for two such subbands whose band index difference $\Delta J = \text{odd}$ ($\Delta J = \text{even}$). Their energy gaps have such a relation: $E_{J_s} - E_{J_s'} \approx (2j - 1)\Delta_{sg}$, where j is an integer. Thus the resonance condition for AGNR can be written as $\Delta_{sg}(N_a) \approx \omega_0/(2j - 1)$. Since intraband (interband) transitions are not permitted for $\Delta J = \text{even}$ ($\Delta J = \text{odd}$), we are finally not able to observe such peaks at $N_a = 84$ and 172 satisfying $\Delta_{sg}(N_a) \approx \omega_0/2j$. Moreover, from p_1 to p_3 , the peaks gradually widen and their cutoff progressively shrinks, as shown in Fig. 2(c). The former can be attributed to the fact that the subband gap is not very sensitive to the change in system size when the size is large enough. The latter stems from the increase in detuning between the driving frequency ω_0 and the excitation gap $(2j - 1)\Delta_{sg}$, deviating from the resonance condition. Analogous to AGNR-HHG, the ZGNR-HHG spectrum shows similar features in Fig. 2(d) and can be explained in the same way.

To gain deeper insights into the mechanism of cutoff extension on the harmonic output, we will study the electron dynamics in the presence of the nearly resonant field. We will show how the electrons are excited to the highest conduction subbands emitting high-energy photons.

III. CASCADE RESONANCE

A. Electron dynamics in GNRs under cascade resonance

Taking 36-ZGNR as an example, we study the HHG process combined with its electron dynamics under resonance. To rule out the effect of pulse envelope on quantum paths, a 10-cycle trapezoid envelope with a one-cycle linear ramp is applied. Figure 3(a) displays the harmonic spectrum when the laser frequency matches the subband gap. It can be seen that the cutoff can reach the 27th order, whereas such a high-order harmonic cannot be observed if the laser is polarized along the ribbon direction. The time-frequency distribution of the corresponding time-dependent current is presented in Fig. 3(b), demonstrating the harmonic emission with subcycle temporal resolution. The maximum order of the emitted harmonics increases with the time evolution at the beginning 3.5 cycles

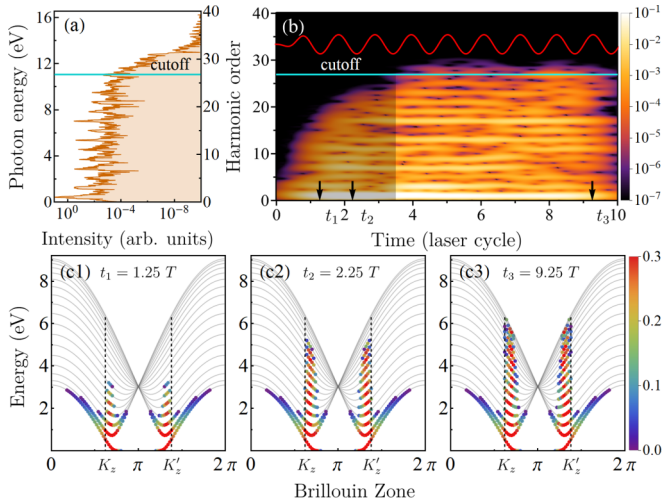


FIG. 3. (a) Harmonic spectrum for 36-ZGNR interacting with a resonant field. The intensity of the harmonic spectrum is in arbitrary units (arb. units). The field strength is $E_0 = 0.0012$ a.u. ($I_0 = 5.04 \times 10^{10}$ W/cm²), and the field wavelength is $\lambda = 3$ μ m ($\omega = 0.0152$ a.u.). (b) Time-frequency distribution of the harmonic emission. The red curve is the vector potential of the laser field. The cyan line indicates the cutoff frequency. (c1)–(c3) Electron dynamics in the conduction subbands for times t_i marked in (b). $T = 2\pi/\omega$ is the optical cycle. The color bar indicates the electron population.

(shadow area) and then stays at 27th order for the following 6.5 cycles. We thus refer to the first 3.5 cycles as the rise stage and the next 6.5 cycles as the oscillation stage.

As shown in Figs. 3(c1)–3(c2), after being excited to the conduction band, the electrons move further up to the higher subbands with increasing vector potential $|A(t)|$. When $|A(t)|$ starts to decrease, however, the electrons near the Dirac points seem not to deexcite to the lower subbands as those electrons governed by Bloch acceleration theorem do. Most of them tend to stay in place and wait for the next half-cycle of the laser pulse. Once $|A(t)|$ increases again, these (deposited) electrons will be excited to the higher subbands. As the electrons are excited and then deposited, accumulation zones are formed gradually near K_z and K'_z . The rise region of the harmonic emission in Fig. 3(b) is exactly ascribed to the cascade excitation process near the Dirac points. When all the subbands are involved (at about $t = 3.5$ T), see Fig. 3(c3), the electrons are no longer excited to the higher subbands, but they jump up and down in the accumulation zones, thus leading to the oscillation region in Fig. 3(b). We thus refer to it as cascade resonance. In AGNR, the resonant dynamics is similar. We provide an animated image of the conduction-band population of 40-AGNR and 36-ZGNR in the Supplemental Material [75] so as to gain more insight into the electron resonant dynamics. Note that the electron dynamics in the valence band is not shown since the distribution of the holes in the valence band is symmetric to the electron distribution on the conduction band.

With a long enough laser pulse, it is not difficult to foresee that the electrons can be driven to the highest subbands, generating high-order harmonics, the energy of which equals the maximum band gap. This explains the significant cutoff

extension on the size-dependent harmonic spectra shown in Figs. 2(c) and 2(d), when the resonance condition is satisfied.

B. Analysis of cascade resonance in simplified model

To analyze the cascade resonance near the Dirac points of GNRs and generalize the conclusions to other confined systems, we introduce an N -level model here that is widely used to simulate electron dynamics and HHG in quantum dots [52,76,77]. The Hamiltonian reads $H = \sum_i^N E_i |i\rangle\langle i|$, where E_i is the energy of state $|i\rangle$. In the dipole approximation, the time-dependent Hamiltonian of the laser field with the N -level quantum dot is written as

$$H(t) = \sum_i^N E_i |i\rangle\langle i| + \mathbf{E}(t) \cdot \sum_{i \neq j} \mathbf{d}_{ij} |i\rangle\langle j|, \quad (1)$$

where electric field $\mathbf{E}(t) = E_0 \cos(\omega t) \hat{y}$, and \mathbf{d}_{ij} is the dipole matrix element between states $|i\rangle$ and $|j\rangle$. Rabi frequency is defined as $\Omega_{ij} = E_0 d_{ij}^y$. In principle, the Hamiltonian Eq. (1) can be used to simulate electron dynamics and HHG in any confined system, including GNRs along the transverse direction, as long as the energy levels and dipole matrix elements are known.

In our simulation, energy levels $E_1, E_2, \dots, E_{N/2}$ are involved as the initial state. Solving the time-dependent Schrödinger equation, the dipole moment can thus be evaluated by $\mathbf{d}(t) = \hat{y} \sum_{i,j} \langle \psi(t) | i \rangle d_{ij}^y \langle j | \psi(t) \rangle$. By the Fourier transformation of $\mathbf{d}(t)$, the harmonic spectrum is obtained, $S(\omega) = |\text{FT}[\mathbf{d}(t)]|^2$.

1. Equispaced N -level model

First of all, we consider an ideal situation: the N -level system with equal separations

$$E_{i+1} - E_i = \Delta_{\text{sg}}, \quad (2)$$

where $i = 1, 2, \dots, N-1$, and Δ_{sg} is the gap between energy levels. The dipole matrix elements (in atomic unit) take the form

$$\begin{aligned} d_{ij} &= 1.0, & |i-j| &= 1, \\ d_{ij} &= 0.3/|E_j - E_i|, & |i-j| &\text{is odd but } > 1, \\ d_{ij} &= 0.0, & |i-j| &\text{is even.} \end{aligned} \quad (3)$$

It is constructed based on the features of dipole matrix elements in GNRs, for example Eq. (B4).

For $N = 2$, the model is reduced to the classical Rabi model, which has been investigated extensively. In the resonant case, $\omega = \Delta_{\text{sg}}$, the parameter

$$\gamma_R = \frac{\Omega_R}{\omega} \quad (4)$$

is commonly adopted to identify different interaction regimes, where $\Omega_R = \Omega_{12} = E_0 |d_{12}|$. In the weak-coupling regime, $\gamma_R \ll 1$, the Rabi flopping can be observed, showing that the population inversion oscillates between -1 and 1 at frequency Ω_R periodically [66], and thus the area theorem is valid [78]. While transforming into the strong-coupling regime, $\gamma_R \gtrsim 1$, the area theorem breaks down [79–81]. A chaotic oscillation mode displaces the periodic mode since the contribution of

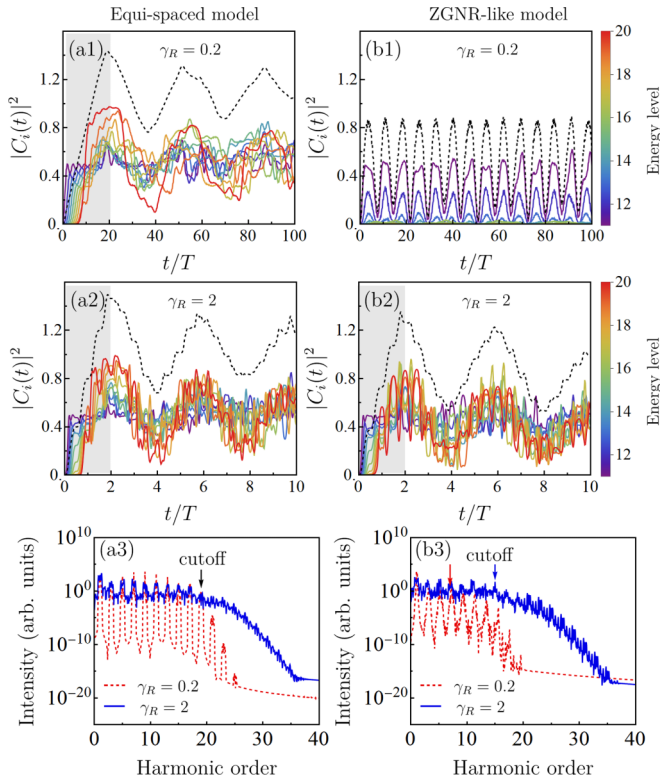


FIG. 4. Population dynamics of conduction states in an equi-spaced model with $N = 20$ in (a1) the weak-coupling regime ($\gamma_R = 0.2$) and (a2) the strong-coupling regime ($\gamma_R = 2$). Corresponding plots for the 20-level ZGNR-like model are shown in (b1) and (b2). The color bar identifies the conduction levels, and the black dashed curve shows the total population of $i = 11, \dots, 20$ scaled by $1/5$ in all cases except (b1). Parts (a3) and (b3) show the HHG spectrum for the equi-spaced and the ZGNR-like systems, respectively. The red and blue curves represent harmonic spectra for $\gamma_R = 0.2$ and 2 , respectively.

counterrotating terms becomes prominent, which is known as the carrier-wave Rabi flopping [82–84].

In a system with $N > 2$ we need to reanalyze its resonant dynamics, because the electron dynamics of the two-level system no longer applies. For comparison, the concepts of weak coupling, strong coupling, and coupling parameter are borrowed. In multilevel systems, we define the coupling parameter as

$$\gamma_R = \frac{\Omega_R}{\omega} = \frac{\langle |d_{ij}| \rangle E_0}{\omega}, \quad (5)$$

where $\langle \dots \rangle$ denotes the average value, d_{ij} is the dipole matrix element between the two energy levels, which meet the resonance condition in frequency, and the Rabi frequency $\Omega_R = \langle |d_{ij}| \rangle E_0$. We focus on the case $\omega = \Delta_{\text{sg}}$ in the following because other resonance cases [$\omega = (2j - 1)\Delta_{\text{sg}}$, $j > 1$] are similar.

Taking $N = 20$ as an example, the resonant dynamics on conduction levels is shown in Figs. 4(a1) and 4(a2). We set the energy separation $\Delta_{\text{sg}} = 1$ a.u., and the driver frequency $\omega = 1$ a.u.. The valence population is not shown because it is symmetric to the conduction population and satisfies $|C_i|^2 + |C_{N+1-i}|^2 = 1$, where $|C_i|^2$ represents the

population on the energy level E_i . In the weak-coupling regime $\gamma_R = 0.2$ ($E_0 = 0.2$ a.u., $\langle d_{i,i+1} \rangle = 1.0$ a.u.), see Fig. 4(a1), the excitation process can be divided into two stages: rise and oscillation, similar to what has occurred in the laser-driving GNR. In the rise region (shadow area), we can find that the electrons are excited to the conduction subbands in a cascaded way. Correspondingly, the total conduction population ($P_c = \sum_{i=N/2+1}^N |C_i|^2$) exhibits a monotonous increase shown by the black dashed line in Fig. 4(a1). Thus the rise time (T_r) is defined as the period from the beginning of the evolution to the first time that P_c reaches the maximum. After entering in the oscillation region, the population of conduction levels starts to collectively oscillate up and down with a roughly uniform period $T_{\text{osc}} \approx 35$ T, where $T = 2\pi/\omega$. Nevertheless, this periodicity is not strict and the population does not return to the initial value even for long enough pulses. Hence the area theorem fails for cascade resonance even in the weak-coupling regime. For strong coupling $\gamma_R = 2$ ($E_0 = 2$ a.u.), the population dynamics is unaltered compared to the case of $\gamma_R = 0.2$ a.u., as shown in Figs. 4(a2). The only significant change is that the oscillation period T_{osc} becomes 3.5 T. Therefore, the regime transformation in the equi-spaced system does not fundamentally shift the behavior of resonant excitation. This is completely different from the two-level resonance.

Based on the above analysis, it is found that the cascade resonance can take place in equi-spaced N -level systems as long as the frequency conditions are met, no matter how weak the electric field is. Because all the energy levels are involved in the cascade resonance, the cutoffs of harmonic spectra can thus extend to the maximum energy gap ($E_{20} - E_1$). Therefore, we can see from Fig. 4(a3) that the harmonic cutoffs reach 19th order [$(E_{20} - E_1)/\omega = 19$] for both $\gamma_R = 0.2$ and 2 , and the intensities of the corresponding harmonic spectra are comparable.

Moreover, the rise time T_r is a continuous function of the Rabi frequency Ω_R and the number of energy levels N . In Appendix C, Figs. 9(a) and 9(b) clearly show that the rise time exhibits a linear dependence of Ω_R^{-1} and N . Mathematically, we describe it by

$$T_r \approx \delta N / \Omega_R = \delta N / (\langle |d_{ij}| \rangle E_0), \quad (6)$$

where δ is the scale factor. This equation indicates that the weaker the field strength is and the more energy levels there are, the longer is the rise time. Reflected in the HHG process of the GNR, this implies that the larger the size and the weaker the field strength, the longer the rise region in harmonic emission.

2. ZGNR-like N -level model

Now we turn to a more realistic situation, taking into account the detuning between the driver frequency and the energy separation. Simulating the subband structure of ZGNR at the Dirac points, energy levels have the form

$$E_{i+1} - E_i = \Delta_{\text{sg}} \left[f - g \left(\frac{i - N/2}{N/2} \right)^2 \right], \quad (7)$$

where f and g are the parameters that determine the nearest-neighbor energy difference. In the following simulation, we

set $N = 20$, $\omega = \Delta_{sg} = 1$ a.u., $f = 1.15$, and $g = 1.1$. The energy difference near the Fermi level is $E_{11} - E_{10} = 1.15 \omega$, slightly deviating from the driver frequency. The dipole matrix elements we used are the same as in Eq. (3).

In this ZGNR-like system, we focus on the population dynamics in different coupling regimes as before. In the weak-coupling regime ($\gamma_R = 0.2$), see Figs. 4(b1), the cascade resonance disappears but is displaced by a behavior close to the classical Rabi flopping. This is called near resonance. Only four of conduction levels near the Fermi level are involved, which implies that only a small proportion of the electrons can be driven to the highest energy level. Therefore, the corresponding HHG spectrum (red dashed curve) in Fig. 4(b3) terminates at seventh order, the energy of which is much lower than the maximum gap $[(E_{20} - E_1)/\omega = 15.37]$. However, in the strong-coupling regime ($\gamma_R = 2$), the collective resonance reappears in the ZGNR-like system, as shown in Fig. 4(b2). The rise and oscillation stages constitute the entire excitation process again. Correspondingly, the cutoff of the harmonic spectrum can thus extend to 15th order, shown as the blue curve in Fig. 4(b3). Therefore, the cascade resonance takes place merely for $\gamma_R \gtrsim 1$ when field detuning exists. This is different from the population dynamics in the equispaced model.

C. Resonance conditions for laser-driving GNRs

Combining the analyses of resonant dynamics in the 36-ZGNR (Sec. III A) and in the simplified models (Sec. III B) together, it is natural to infer that one of the conditions for the occurrence of cascade resonance in the laser-driving GNRs is $\gamma_R \gtrsim 1$. We verify the coupling parameter at p_1 in the ZGNR-HHG spectrum [Fig. 2(d)], and we find that $\gamma_R(N_z = 36) \approx 1.3 > 1$ at the Dirac points.

Thereby, the resonance conditions for a laser-driving GNR can be summarized as

$$\Delta_{sg}(N) \approx \omega/(2j - 1), \quad (8a)$$

$$\gamma_R(N) = E_0 \langle |d_{\Delta J}^y(N)| \rangle / \omega \gtrsim 1, \quad (8b)$$

where j is an integer, and ΔJ is the index difference of resonant subbands. Equation (8a) is the frequency condition we have obtained in Sec. II A. It determines which driver frequency can lead to cascade resonance in a GNR with a size of N . Then Eq. (8b) states that the occurrence of cascade resonance requires the laser-driving GNR to be in the strong-coupling regime, and $\gamma_R = 1$ is the critical point. Further, we define a concept—critical field strength, as

$$E_{\text{cri}}(\Delta J, N) = \omega / \langle |d_{\Delta J}^y(N)| \rangle, \quad (9)$$

which is the minimum field strength resulting in the cascade resonance in N -GNR. Substituting Eq. (8a) into Eq. (9), the critical field strength can be evaluated by

$$E_{\text{cri}}(\Delta J, N) = (2j - 1)\Delta_{sg}(N) / \langle |d_{\Delta J}^y(N)| \rangle. \quad (10)$$

When $j = 1$, Δ_{sg} including Δ_{sg}^{max} and Δ_{sg}^{ave} is a linear function of N^{-1} [Figs. 2(a) and 2(b)], and $\langle |d_{\Delta J=1}^y(N)| \rangle$ is linear with N (Fig. 8). Substituting the linear fitting results of Δ_{sg}^{max} and $\langle |d_{\Delta J=1}^y(N)| \rangle$ into Eq. (10), we finally obtain $E_{\text{cri}}(N)$ as a

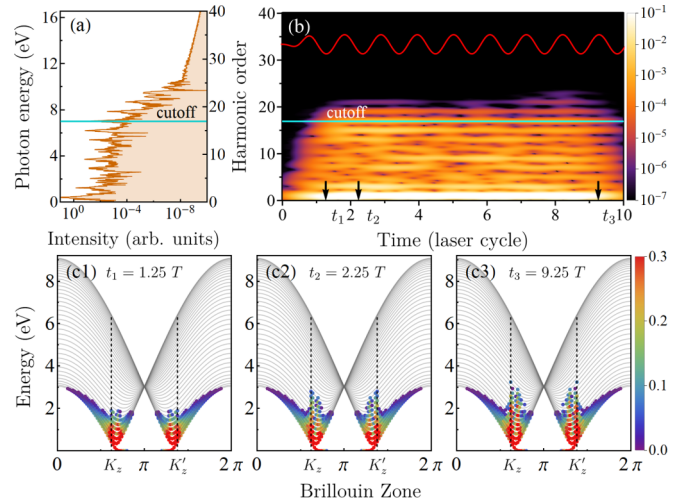


FIG. 5. (a) Harmonic spectrum for 80-ZGNR. The field strength is $E_0 = 0.0012$ a.u. ($I_0 = 5.04 \times 10^{10}$ W/cm²), and the field wavelength is $\lambda = 3 \mu\text{m}$ ($\omega = 0.0152$ a.u.). (b) Time frequency distribution of the harmonic emission. The red curve is the vector potential of the external field. The cyan line indicates the cutoff frequency. (c1)–(c3) Electron dynamics in the conduction subbands for the times t_i marked in (b).

function of N ,

$$\begin{aligned} E_{\text{cri}}^a(N_a) &= (\alpha_a N_a^2 + \beta_a N_a + \gamma_a)^{-1}, & \text{AGNR}, \\ E_{\text{cri}}^z(N_z) &= (\alpha_z N_z^2 + \beta_z N_z + \gamma_z)^{-1}, & \text{ZGNR}, \end{aligned} \quad (11)$$

where $(\alpha_a, \beta_a, \gamma_a) \approx (0.025, 0.039, 0.014)$, and $(\alpha_z, \beta_z, \gamma_z) \approx (0.021, 0.04, 0.014)$. It exhibits a dependence of $E_{\text{cri}}(N) \sim N^{-2}$ in the large- N limit. It clearly indicates that the cascade resonance induced harmonic cutoff extension is more easily observed in large size GNRs.

D. Electron dynamics and HHG away from the resonance condition in GNRs

A natural question at this stage is how sensitive is the cascade resonance to the frequency-matching and field-strength conditions.

To examine the frequency-matching condition, we choose the 80-ZGNR ($\Delta_{sg} \approx 0.2$ eV), but we use the laser field with a wavelength of $3 \mu\text{m}$ ($\omega \approx 0.41$ eV), which causes cascade resonance in 36-ZGNR ($N_z = 36$) to study the HHG process. In Figs. 5(a) and 5(b), the plateau of the harmonic spectrum merely extends to 17th order, which is evidently shorter than that in 36-ZGNR (27th order). In Figs. 5(c1)–5(c3), it is found that the electron dynamics is far from the cascade resonance. On the subbands with energies above 3.5 eV, the population is not as significant as in the case of a cascade resonance. This is because the relation between subband gap and laser frequency ($\Delta_{sg} \approx \omega/2$) does not agree with the frequency condition in Eq. (8a). Nevertheless, the subband gaps are not uniform. The gaps near the Fermi energy are larger than those that are higher, which can satisfy the frequency condition to some extent. Therefore, we observe that subbands close to the Fermi energy are involved in a near resonance. The cascade resonance for 80-ZGNR, however, appears if we employ a laser with wavelength $\lambda = 6 \mu\text{m}$ ($\omega \approx 0.21$ eV) satisfying the

frequency condition ($\Delta_{\text{sg}} \approx \omega$). The accumulation zones of electrons span from 0 to 6.5 eV, and a harmonic cutoff exceeds 11 eV (not shown).

When the laser field does not reach the threshold strength [Eq. (10)], the electron dynamics in GNRs is similar to what has been explored in the ZGNR-like model; see Sec. III B. The electrons are not able to be excited to high-energy subbands, such as that presented in Figs. 5(c1)–5(c3). The intensity of the harmonic spectrum is lower and the plateau is shorter than those in the strong laser field. In the next section, the study of cutoff law on the field strength systematically shows the variation of the harmonic spectrum from the weak-coupling regime to the strong-coupling regime.

IV. CUTOFF LAW OF HHG IN GRAPHENE NANORIBBONS

The cutoff law of HHG is a fundamental issue in strong-field physics. In GNRs, it has been shown that the energy cutoff increases linearly with increasing field strength and wavelength for fields along the ribbon direction, as also shown in Appendix D. This is consistent with the generic observations in bulk solids. When the laser polarization turns to the cross-ribbon direction, however, the accelerated motion of electrons on the energy bands switches to the electron dynamics for finite systems like cascade resonance, thus leading to the alteration of the cutoff law as well. In the following, we explore the cutoff law of GNR-HHG on the laser duration, field strength, and wavelength.

Duration. Figure 6(a) shows the HHG spectra in 80-AGNR subjected to resonant laser pulses with different durations. In the calculation, laser pulses in the form of Eq. (A10) are employed, so the number of laser cycles (n_{cyc}) can be used to indicate pulse duration. We note that the plateau of the harmonic spectrum is gradually stretched with increasing laser duration. In Fig. 6(b), the harmonic cutoff clearly exhibits linear scaling with pulse duration for $n_{\text{cyc}} \leq 5$, and it reaches the maximum attainable energy at $n_{\text{cyc}} = 11$. Between $n_{\text{cyc}} = 6$ and 10, the cutoff varies slowly with increasing laser duration. This cutoff law can be interpreted qualitatively by the two-stage excitation process in GNRs under the cascade resonance, as elaborated in Secs. III A and III B. The linear increase and the slow variation of the cutoff corresponds to the rise and oscillation stages in the resonant excitation, respectively.

Field strength. In Fig. 6(c), we study the field strength dependence of harmonic emissions in 40-AGNR interacting with resonant pulses. The blue line represents the critical field strength for 40-AGNR in which $E_{\text{cri}} = 8 \times 10^{-4}$ a.u. (i.e., $I_{\text{cri}} = 2.24 \times 10^{10}$ W/cm²), and the red line is located at the field strength ($E_{\text{sec}} = 4 \times 10^{-4}$ a.u.) in which the harmonic spectrum starts to possess a marked second plateau. Given these two lines, the spectrum is intuitively divided into three regions: (i) $\gamma_R \ll 1$, (ii) $\gamma_R \lesssim 1$, and (iii) $\gamma_R > 1$. (i) In the weak-coupling regime ($\gamma_R \ll 1$), the energy cutoff presents a linear dependence on the field strength; see Figs. 6(c) and 6(d). (ii) When entering the transition regime ($\gamma_R \lesssim 1$), we can observe two-plateau structures on HHG spectra, such as the red dashed curve in Fig. 6(e), which result from the difference of population on lower and higher subbands. This difference can be understood by the similar electron dynamics in

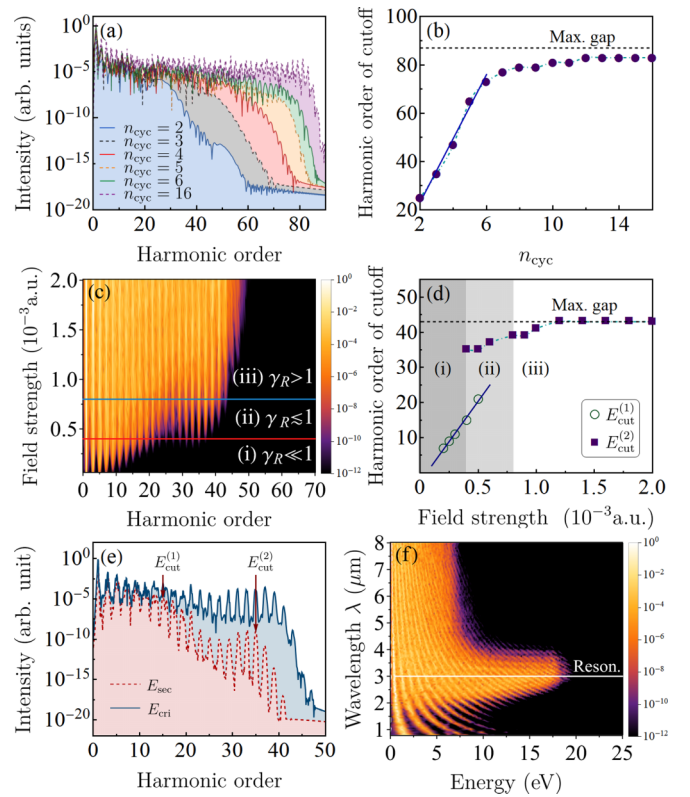


FIG. 6. (a) Harmonic spectra vs number of cycles (n_{cyc}) in 80-AGNR. The field strength is $E_0 = 5 \times 10^{-4}$ a.u. ($I_0 = 8.75 \times 10^9$ W/cm²), $\gamma_R \approx 1.25$, and the frequency is $\omega_0 = 0.0076$ a.u. ($\lambda = 6 \mu\text{m}$). (b) The cutoff as a function of n_{cyc} corresponding to the harmonic spectra in (a). (c) Harmonic spectra vs field strength in 40-AGNR, using a driver with $n_{\text{cyc}} = 16$, and $\omega_0 = 0.0152$ a.u. ($\lambda = 3 \mu\text{m}$). The color bar indicates the intensity of harmonics. (d) The cutoff as a function of field strength, corresponding to the harmonic spectra in (c). (e) Harmonic spectra extracted from (c) for $E_{\text{sec}} = 4 \times 10^{-4}$ a.u. and $E_{\text{cri}} = 8 \times 10^{-4}$ a.u. (f) Harmonic spectra vs wavelength in 40-AGNR, using a driver with $n_{\text{cyc}} = 16$, and $E_0 = 0.0012$ a.u. ($I_0 = 5.04 \times 10^{10}$ W/cm²). The white line represents the onset of cascade resonance.

the ZGNR-like system, that is, the lower subbands participate in collective resonance, whereas the higher subbands are in near resonance or even in nonresonance. As Fig. 6(d) shows, the cutoff of the first plateau ($E_{\text{cut}}^{(1)}$) preserves the linear dependence in region (i), but the second cutoff ($E_{\text{cut}}^{(2)}$) approaches the attainable maximum tardily. (iii) As the field strength increases up to E_{cri} , the blue curve in Fig. 6(e) shows that the first and the second plateaus merge together since all subbands are involved in the collective resonance. For this reason, the energy cutoff in the strong-coupling regime ($\gamma_R > 1$) tends to saturate and remains at the maximum band gap. We have thus revealed that the cutoff law of HHG in GNRs depends strongly on the interaction regime to which the system is subjected, the cutoff neither following the linear scaling in the electric field of the bulk solids nor the quadratic dependence in the electric field of gas.

Wavelength. Figure 6(f) shows the HHG spectrum as a function of driver wavelength in 40-AGNR ($\Delta_{\text{sg}} \approx 0.4$ eV). It can be found that the cutoff frequency is extended abruptly

around a wavelength $\lambda = 3 \mu\text{m}$ ($\omega \approx 0.41 \text{ eV}$), where the frequency condition is satisfied exactly. This is because the cascade resonance appears as the wavelength approaches $3 \mu\text{m}$. When the driver wavelength increases away from $3 \mu\text{m}$, the laser-driving AGNR begins to deviate from the resonance point, and thus the plateau of the harmonic spectrum shrinks drastically.

V. DISCUSSION OF CASCADE RESONANCE IN GENERAL CONFINED SYSTEMS

We now turn our attention to the possibility of observing the cascade resonance induced HHG spectra in general confined systems. The key ingredients for cascade resonance, viz., the frequency-matching and field strength conditions, can be approximately met in a variety of systems in a nanoribbon geometry [59], for example in the kagomé lattice [85] and $\alpha\text{-T}_3$ lattice [86,87] systems. Additionally, quantum dots offer a high degree of tunability in terms of the energy spectra via the synthesis route, size dependence, and/or the external parameters. In general, however, for most materials/systems, not only is the frequency-matching condition over a large number of subbands potentially not possible, but also the fundamental gap (E_g) could be significantly different from the (average) subband gap (Δ_{sg}).

To explore such systems for possible cascade resonance induced HHG phenomena, we resort to the simplified models akin to the ones introduced in Sec. III B, but the dipole matrix element takes the form

$$\begin{aligned} d_{ij} &= 1.0/|E_j - E_i|, & |i - j| \text{ is odd,} \\ d_{ij} &= 0.0, & |i - j| \text{ is even.} \end{aligned} \quad (12)$$

We consider $E_g \neq \Delta_{\text{sg}}$ and laser frequency matched to either the fundamental gap or the average sub(band) gap. Therefore, we need two parameters,

$$\gamma_E = d_{E_g} E_0 / \omega = E_0 / (\omega E_g), \quad (13)$$

$$\gamma_\Delta = \langle d_{i,i+1} \rangle E_0 / \omega = E_0 / (\omega \langle \Delta_{\text{sg}} \rangle) \quad (14)$$

denoting the coupling strength for the fundamental gap and the sub(band) gap, respectively. We additionally consider cases in which Δ_{sg} is nonuniform. To illustrate these cases, we also consider a 20-level model.

Case I: $E_g < \Delta_{\text{sg}}$. For brevity, we consider the fundamental gap $E_g = 1 \text{ a.u.}$ while all other energy levels are equally spaced in energy with a larger gap:

$$E_{i+1} - E_i = E_g = 1 \text{ a.u.} \quad \text{for } i = N/2, \quad (15a)$$

$$E_{i+1} - E_i = \Delta_{\text{sg}} = 4 \text{ a.u.} \quad \text{otherwise.} \quad (15b)$$

When the laser frequency matches the fundamental gap E_g , the system exhibits near resonance irrespective of the coupling strength. The electron dynamics in the strong coupling ($E_0 = 2 \text{ a.u.}$, $\gamma_E = 2$) is shown in Fig. 7(a1). Significant electron population and oscillation merely occur at the energy levels close to the Fermi energy. On the other hand, when the laser frequency matches the sub(band) gap Δ_{sg} , the cascade resonance can be achieved, in principle, even for the weak coupling between the sub(band) levels, as shown in Fig. 7(a2)

for $\gamma_\Delta = 0.125$ ($E_0 = 2 \text{ a.u.}$). The population on each energy level is comparable.

The corresponding HHG spectra for different tuning of the laser frequency are shown in Fig. 7(a3). We clearly see that the HHG spectrum for the cascade resonance when $\omega = \Delta_{\text{sg}}$ is much more intense than for the near resonance when $\omega = E_g$. The frequency cutoff of the resonant harmonic spectrum reaches 72 a.u., which is close to the maximum band gap (73 a.u.).

Case II: $E_g > \Delta_{\text{sg}}$. We set the fundamental gap and the sub(band) gap as

$$E_{i+1} - E_i = E_g = 4 \text{ a.u.} \quad \text{for } i = N/2, \quad (16a)$$

$$E_{i+1} - E_i = \Delta_{\text{sg}} = 1 \text{ a.u.} \quad \text{otherwise.} \quad (16b)$$

Figure 7(b1) shows the population dynamics for the laser frequency matching the fundamental gap E_g . The field strength is set by $E_0 = 1.0 \text{ a.u.}$ ($\gamma_E = 0.625$). The near resonance which has been observed in Fig. 7(a1) occurs in the energy levels near the Fermi level. Therefore, in the corresponding HHG spectrum, see Fig. 7(b3), the intensity is relatively lower and the plateau structure is not clear. On the other hand, when the laser frequency matches the sub(band) gap, a cascade resonance can occur only when $\gamma_\Delta \gtrsim 1$. Figure 7(b2) shows the electron dynamics when $E_0 = 2 \text{ a.u.}$ ($\gamma_\Delta = 2$). The cascade resonance takes place. The harmonic cutoff, blue curve in Fig. 7(b3), can thus reach the maximum energy gap at 22 a.u. ($E_{20} - E_1$).

We further note that the cascade resonance depends crucially on the ratio of E_g / Δ_{sg} . For much larger fundamental gap values, the electron density in the higher subbands decreases as the excitation of electrons to the first conduction level is probabilistically low. The cascade resonance then cannot take place. In the specific context of GNRs, this can be achieved by adding staggered on-site potentials which open or enlarge the fundamental gap. In Appendix E, we show the HHG spectrum and population dynamics in 36-ZGNR with a 1.0 eV gap. We can still observe the cascade resonance near the Dirac points, but there is a lower conduction population in comparison to that of gapless 36-ZGNR. The intensity of the corresponding HHG spectrum is lower than that of the gapless ZGNR-HHG spectrum. However, the cascade resonance disappears when the fundamental gap is larger than 1.5 eV (not shown).

A more realistic scenario in materials is when the sub(band) gaps are nonuniform. To account for these effects, we consider a model where Δ_{sg} is chosen randomly (uniform probability distribution) in a given energy range, while $E_g > \langle \Delta_{\text{sg}} \rangle$, where $\langle \dots \rangle$ denotes the average value. The energy levels are

$$E_{i+1} - E_i = E_g = 4 \text{ a.u.} \quad \text{if } i = N/2, \quad (17a)$$

$$E_{i+1} - E_i = \Delta_{\text{sg}} = \Delta \text{Ran}(0.5, 1.5) \quad \text{otherwise,} \quad (17b)$$

where $\text{Ran}(n, m)$ means a random number between n and m . The laser frequency matches the average sub(band) gap ($\omega = \Delta = 1 \text{ a.u.}$).

Figure 7(c1) shows the electron dynamics in the weak-coupling regime, where $E_0 = 0.2 \text{ a.u.}$ ($\gamma_\Delta \approx 0.2$). Because of the large fundamental gap ($E_g = 4 \text{ a.u.}$), the electrons can hardly be excited to conduction levels. The population of the

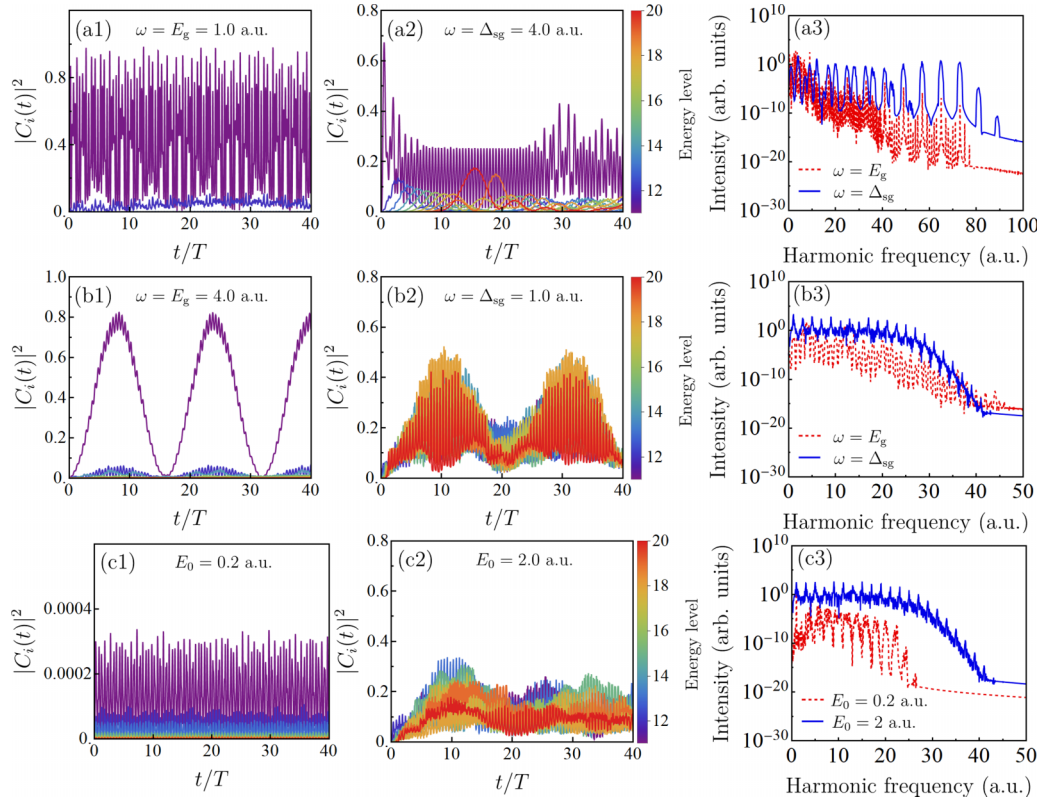


FIG. 7. Population dynamics and HHG spectrum depending on energy-level structure. (a1),(a2) Population dynamics for $E_g < \Delta_{sg}$. (a1) Laser frequency matches fundamental gap ($\omega = E_g = 1$ a.u.). (a2) Laser frequency matches sub(band) gap ($\omega = \Delta_{sg} = 4$ a.u.). (a3) Harmonic spectra in red and blue correspond to the electron dynamics in (a1) and (a2), respectively. (b1),(b2) Population dynamics for $E_g > \Delta_{sg}$. (b1) Laser frequency matches fundamental gap ($\omega = E_g = 4$ a.u.). (b2) Laser frequency matches sub(band) gap ($\omega = \Delta_{sg} = 1$ a.u.). (b3) Harmonic spectra in red and blue correspond to the electron dynamics in (b1) and (b2), respectively. (c1),(c2) Population dynamics for a system with random sub(band) gap in (c1) the weak-coupling regime ($E_0 = 0.2$ a.u.) and (c2) the strong-coupling regime ($E_0 = 2$ a.u.). The laser frequency matches the average sub(band) gap. (c3) Harmonic spectra in red and blue correspond to the electron dynamics in (c1) and (c2), respectively.

first conduction level is even less than 0.01. In comparison, in the strong-coupling regime ($E_0 = 2$ a.u.), shown in Fig. 7(c2), all the higher-lying energy levels have a sizable population. The details of the energy-level distribution seem immaterial to the cascade resonance phenomena. These features are also clearly reflected in the HHG spectra. In Fig. 7(c3), the intensity of the harmonic spectrum in the strong-coupling regime is much higher than that in the weak-coupling regime.

Consequently, the cascade resonance can be anticipated in real nanoribbon materials where a few subbands are nearly equispaced in energy while other bands are randomly distributed. When the laser frequency matches the subband gap, the resonant excitation allows more electrons to occupy higher conduction subbands, which eventually enhances and broadens the harmonic spectra in experimental measurements.

VI. CONCLUSION AND OUTLOOK

In conclusion, we demonstrated theoretically that the cascade resonance provides a systematic way to extend and enhance the harmonic spectrum of a confined quantum system. The cascade resonance can pump electrons to higher subbands, resulting in enhanced higher-order harmonic emissions, which is, therefore, fundamentally distinct from the

high-harmonic generation mechanism of atoms, molecules, and bulk.

Based on the study of size-dependent GNR-HHG and dynamic analysis in a multilevel model, the resonance conditions for laser parameters such as frequency and field strength are established. The cascade resonance occurs when the laser frequency matches the subband gap and the field exceeds the threshold strength. While large deviations from the ideal frequency-matching condition lead to the disappearance of the cascade resonance, for small deviations strong field strengths can still induce cascade resonance, although the strength of the HHG spectra becomes relatively weak.

These predictions are well within experimental reach, and they are relatively straightforward to verify in the present experimental setups. These ideas can also be applicable to other confined materials/systems that, in general, may not meet the frequency-matching condition perfectly. Perhaps the most interesting situation is when only a part of the subbands meets the frequency-matching condition, where the cascade resonance of a few levels may take place by carefully tuning the laser frequency and intensity, for example in other two-dimensional materials in a stable nanoribbon geometry, or in quantum dots. Among these, nanoribbons based on the kagomé lattice and α - T_3 lattice systems are particularly appealing due to the presence of a flat band together with the

obtaining the eigenstates ϕ_{nk_x} and eigenvalues $E_n(k_x)$ of the system. In Figs. 1(c) and 1(d), we show the energy bands of AGNR and ZGNR in the case of $N_a = 10$ and $N_z = 10$, which are calculated by plugging Eqs. (A6) and (A7) into Eq. (A8), respectively.

2. Coupling to an external laser field and numerical calculations

To simulate the interaction of a laser field and GNR, we use the time-dependent Schrödinger equation

$$i \frac{\partial}{\partial t} \Psi(t) = \mathbf{H}(t) \Psi(t), \quad (\text{A9})$$

where $\mathbf{H}(t)$ is the matrix form of the time-dependent Hamiltonian, which has been coupled to the external field $\mathbf{A}(t)$.

Here, the vector potential $\mathbf{A}(t) = (0, A_y)^T$ reads

$$A_y(t) = \frac{E_0}{\omega} \sin^2 \left(\frac{\omega t}{2n_{\text{cyc}}} \right) \sin(\omega t) \quad (\text{A10})$$

in the domain $0 \leq t \leq n_{\text{cyc}}T$, where E_0 is the amplitude of the electric field, ω is the fundamental frequency, $T = 2\pi/\omega$ is the time period, and n_{cyc} is the total number of laser cycles.

For a tight-binding GNR, the driving fields shift the quasi-momentum k_x like

$$k_x \rightarrow k_x(t) = k_x + A_x(t), \quad (\text{A11})$$

because of the periodic boundary condition in the x direction [88]. The y component of the drivers will induce phase factors on the hopping terms, which makes the replacement of

$$\gamma_{ij} \rightarrow \gamma_{ij} e^{-iA_y(t)(y_j - y_i)}, \quad (\text{A12})$$

where γ_{ij} represents the hopping integral between site i and site j [88].

Thereby, according to the above statement, the matrix elements of the time-dependent AGNR Hamiltonian $\mathbf{H}^a(t)$ are expressed by

$$\begin{aligned} & \mathbf{H}_{i,i+1}^a[k_x(t)], & i = 1, 2, \dots, 2N_a - 1, \\ & \gamma_y^a(t) \cdot \mathbf{H}_{i,i+2}^a[k_x(t)], & i = 1, 2, \dots, 2N_a - 2, \\ & 0, & \text{otherwise,} \end{aligned} \quad (\text{A13})$$

where the phase factor $\gamma_y^a(t) = e^{iaA_y(t)/2}$ and $\mathbf{H}_{ij}^a(k_x)$ correspond to the elements in Eq. (A6). Similarly, the elements of ZGNR Hamiltonian $\mathbf{H}^z(t)$ read

$$\begin{aligned} & \gamma_{y,1}^z \cdot \mathbf{H}_{2i-1,2i}^z[k_x(t)], & i = 1, 2, \dots, N_z/2, \\ & \gamma_{y,1}^z \cdot \mathbf{H}_{2i,2i+1}^z[k_x(t)], & i = 1, 2, \dots, N_z/2 - 1, \\ & 0 & \text{otherwise,} \end{aligned} \quad (\text{A14})$$

where $\gamma_{y,1}^z(t) = e^{i\frac{aA_y}{2\sqrt{3}}}$, $\gamma_{y,2}^z(t) = e^{i\frac{aA_y}{\sqrt{3}}}$, and $\mathbf{H}_{ij}^z(k_x)$ correspond to the elements in Eq. (A7). It is important to note that only the upper triangular elements of $\mathbf{H}(t)$ are given in Eqs. (A13) and (A14), and one can easily obtain the values of lower triangular elements by using $\mathbf{H}_{ji} = (\mathbf{H}_{ij})^*$.

In this simulation, the initial states $\Psi_{nk_x}(t=0)$ are determined by the lowest $N/2$ eigenstates ϕ_{nk_x} from Eq. (A8), where N indicates the total number of eigenstates. The wave functions are numerically propagated with the Crank-Nicolson method [89]:

$$\Psi(t + \Delta t) \simeq \left[\mathbf{I} - \frac{\mathbf{H}(t)\Delta t}{2i} \right]^{-1} \left[\mathbf{I} + \frac{\mathbf{H}(t)\Delta t}{2i} \right] \Psi(t), \quad (\text{A15})$$

where \mathbf{I} is the identity matrix, and Δt is the discrete time step for temporal evolution. Then we can calculate the value of induced electric current as

$$J_\alpha(t) = - \sum_{n=1}^{N/2} \sum_{k_x \in \text{BZ}} \Psi_{nk_x}^\dagger(t) \left(\frac{\partial \mathbf{H}}{\partial A_\alpha} \right) \Psi_{nk_x}(t), \quad (\text{A16})$$

where n labels the state, N is the size of the Hamiltonian matrix, $\alpha = x, y$, and $\Psi_{nk_x}^\dagger = (\Psi_{nk_x}^*)^T$. Ultimately, the harmonic spectrum along the α direction can be evaluated from the current by

$$S_\alpha(\omega) = \left| \text{FT} \left[\frac{d}{dt} J_\alpha(t) \right] \right|^2. \quad (\text{A17})$$

Note that the derivative of current is multiplied by a Blackman window before the Fourier transform.

APPENDIX B: OPTICAL SELECTION RULES IN GNRs

Since the selection rules of GNR subbands [90–94] are crucial for understanding the size-dependent HHG spectra in Sec. II A, it is necessary for us to clarify them in this Appendix.

In general, the dipole matrix element of eigenstates ϕ_a and ϕ_b is written as

$$d_{ab}^\alpha = \langle \phi_a | \hat{r}_\alpha | \phi_b \rangle = -i \frac{\langle \phi_a | \hat{p}_\alpha | \phi_b \rangle}{E_a - E_b}, \quad (\text{B1})$$

where \hat{r}_α is the position operator along the α direction, \hat{p}_α is the momentum operator along the α direction, and E_a and E_b are the eigenenergies of states ϕ_a and ϕ_b . In the nanoribbon model, the eigenvector ϕ_{nk_x} is just a function of k_x and y because the Fourier transformation is made in the x direction. Thus the dipole matrix elements between subbands $J_1(s)$ and $J_2(s')$ are written as

$$d_{J_1s, J_2s'}^x(k_x) = i \phi_{J_1s}^{s\dagger} \partial_{k_x} \phi_{J_2s'}^s = -i \frac{\phi_{J_1s}^{s\dagger} \frac{\partial \mathbf{H}}{\partial k_x} \phi_{J_2s'}^s}{E_{J_1} - E_{J_2}}, \quad (\text{B2})$$

$$d_{J_1s, J_2s'}^y(k_x) = \phi_{J_1s}^{s\dagger} \mathbf{y} \phi_{J_2s'}^s = -i \frac{\phi_{J_1s}^{s\dagger} \mathbf{p}_y \phi_{J_2s'}^s}{E_{J_1} - E_{J_2}}, \quad (\text{B3})$$

where s and s' are the indices of band type, J_1 and J_2 are the subband numbers, E_J is the band dispersion of subband J , ϕ_J^s is the eigenvector obtained from Eq. (A8), $\mathbf{y} = \text{diag}\{y_1, y_2, \dots, y_N\}$ is the matrix form of the position operator in the y direction, y_1, y_2, \dots, y_N are the coordinates of atom sites on the y axis, and $\mathbf{p}_y = -i[\mathbf{y}, \mathbf{H}]$.

In what follows, only the y component of the dipole matrix element $d_{J_1s, J_2s'}^y$ is discussed, because we focus mainly on the optical transition while the GNR is driven by y -polarized light. In Eq. (B4), we give an example of a dipole matrix (modulus)

of 10-ZGNR at Dirac point K_z ,

$$|d_{J_1 s, J_2 s'}^y(K_z)| = \begin{pmatrix} 0.0 & 3.99 & 0.0 & 0.32 & 0.0 & 0.084 & 0.0 & 0.03 & 0.0 & 0.33 \\ 3.99 & 0.0 & 4.30 & 0.0 & 0.40 & 0.0 & 0.113 & 0.0 & 0.297 & 0.0 \\ 0.0 & 4.30 & 0.0 & 4.39 & 0.0 & 0.43 & 0.0 & 0.213 & 0.0 & 0.03 \\ 0.32 & 0.0 & 4.39 & 0.0 & 4.42 & 0.0 & 0.103 & 0.0 & 0.114 & 0.0 \\ 0.0 & 0.40 & 0.0 & 4.42 & 0.0 & 4.09 & 0.0 & 0.43 & 0.0 & 0.084 \\ 0.084 & 0.0 & 0.43 & 0.0 & 4.09 & 0.0 & 4.42 & 0.0 & 0.40 & 0.0 \\ 0.0 & 0.113 & 0.0 & 0.103 & 0.0 & 4.42 & 0.0 & 4.39 & 0.0 & 0.32 \\ 0.03 & 0.0 & 0.213 & 0.0 & 0.43 & 0.0 & 4.39 & 0.0 & 4.30 & 0.0 \\ 0.0 & 0.297 & 0.0 & 0.114 & 0.0 & 0.40 & 0.0 & 4.30 & 0.0 & 3.99 \\ 0.33 & 0.0 & 0.03 & 0.0 & 0.084 & 0.0 & 0.32 & 0.0 & 3.99 & 0.0 \end{pmatrix}, \quad (\text{B4})$$

in the basis $(|5v\rangle, |4v\rangle, \dots, |1v\rangle, |1c\rangle, \dots, |5c\rangle)^T$. We can see some zero matrix elements that result in the interesting selection rule in ZGNRs.

First, let us examine the selection rule in AGNRs. According to the analytical derivations in Refs. [92–94] and our numerical calculations, the dipole matrix elements of the intraband from the same group, i.e., $s' = s$, and $(J_1, J_2) \in \{J\}$ or $\{J'\}$, possess the following feature:

$$\begin{aligned} d_{J_1 s, J_2 s}^y &= 0, & \Delta J &= J_2 - J_1 \in \text{even}, \\ d_{J_1 s, J_2 s}^y &\neq 0, & \Delta J &= J_2 - J_1 \in \text{odd}. \end{aligned} \quad (\text{B5})$$

For the interband transition ($s \neq s'$), dipole matrix elements from the same group show

$$\begin{aligned} d_{J_1 s, J_2 s'}^y &\neq 0, & \Delta J &= J_2 - J_1 \in \text{even}, \\ d_{J_1 s, J_2 s'}^y &= 0, & \Delta J &= J_2 - J_1 \in \text{odd}. \end{aligned} \quad (\text{B6})$$

That is to say, the intraband (interband) transition between those two subbands is forbidden whenever the difference in the corresponding indices (ΔJ) is an even (odd) number. Then let us move to another case, considering the transition between the subbands in different groups, that is, $J_1 \in \{J\}$ and $J_2 \in \{J'\}$. The intraband ($s = s'$) dipole moments are shown as

$$\begin{aligned} d_{J_1 s, J_2 s}^y &\neq 0, & \Delta J &= J_2' - J_1 \in \text{even}, \\ d_{J_1 s, J_2 s}^y &= 0, & \Delta J &= J_2' - J_1 \in \text{odd}. \end{aligned} \quad (\text{B7})$$

For the interband transition ($s \neq s'$), the situation is just opposite to Eq. (B7), that is, the optical transition is forbidden whenever the index difference ΔJ is even, shown as

$$\begin{aligned} d_{J_1 s, J_2 s'}^y &= 0, & \Delta J &= J_2' - J_1 \in \text{even}, \\ d_{J_1 s, J_2 s'}^y &\neq 0, & \Delta J &= J_2' - J_1 \in \text{odd}. \end{aligned} \quad (\text{B8})$$

Secondly, we consider the selection rule of ZGNR, which is simpler than that of AGNR, because the ZGNR only has one group of subbands. Based on a similar calculation and analysis, we find that the features of dipole matrix elements for intraband and interband transitions are just the same as that shown in Eqs. (B5) and (B6). The intraband transition is allowed when the index difference ΔJ is odd, whereas the interband transition is allowed when the index difference ΔJ is even.

Focusing on the intraband transitions near the Dirac points, we compute the dipole matrix elements $d_{J, J+1}^y$ for AGNR and ZGNR of nearest-neighbor subbands at Dirac points using Eq. (B3). It shows an interesting characteristic that the moduli of $d_{J, J+1}^y(K)$ are almost the same with different band

numbers J , for example $d_{\Delta J=1}^y(K_z)$ in Eq. (B4). So we plot $\langle |d_{\Delta J=1}^y(K_a)| \rangle$ and $\langle |d_{\Delta J=1}^y(K_z)| \rangle$, the average values of dipole matrix elements of nearest-neighbor subbands at Dirac points, in Figs. 8(a) and 8(b), respectively. For AGNR, the results of matrix elements from the second group $\{J'\}$ are not presented in Fig. 8(a). We can see clearly that the dipole matrix elements $\langle |d_{\Delta J=1}^y| \rangle$, for both AGNR and ZGNR, vary linearly with the number of sites:

$$\langle |d_{\Delta J=1}^y(K_a)| \rangle = 0.47N_a + 0.26, \quad (\text{B9a})$$

$$\langle |d_{\Delta J=1}^y(K_z)| \rangle = 0.41N_z + 0.20. \quad (\text{B9b})$$

This indicates that the coupling between nearest-neighbor subbands becomes stronger while increasing the ribbon width.

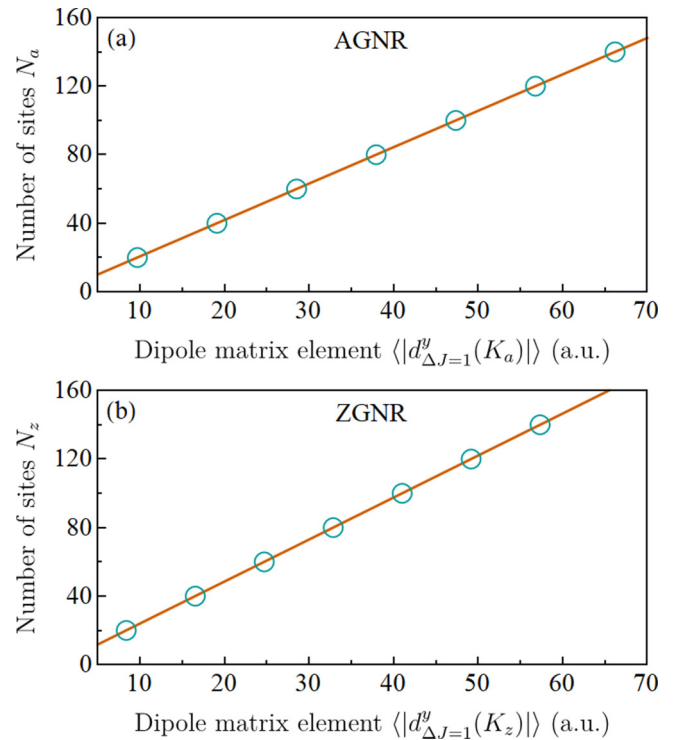


FIG. 8. Dipole matrix elements $\langle |d_{\Delta J=1}^y(K_l)| \rangle$ with respect to the ribbon widths N_l for (a) AGNRs ($l = a$) and (b) ZGNRs ($l = z$). The open circles are data points, and orange lines are the linear fit of the data points.

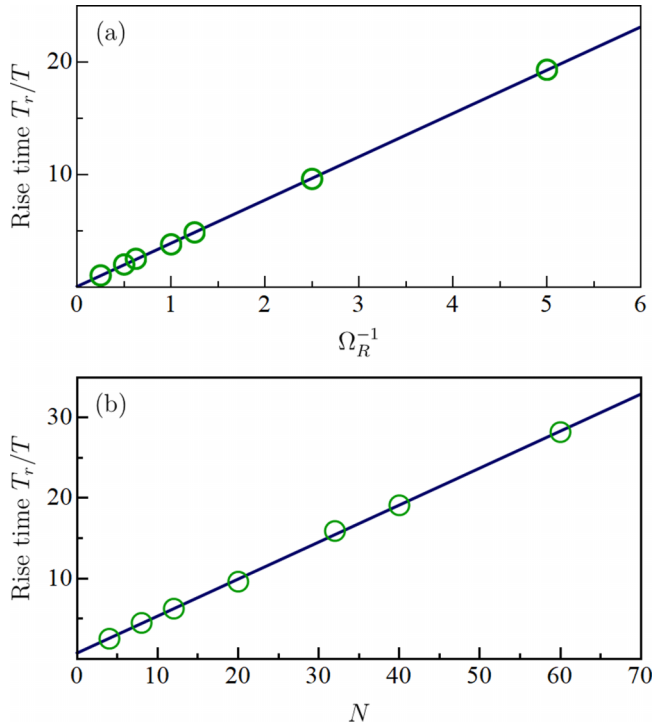


FIG. 9. (a) Linear rise time as a function of Ω_R^{-1} for $N = 20$. The dipole matrix elements are the same as in Eq. (3). Laser frequency is set to $\omega = \Delta_{\text{sg}} = 1$ a.u. (b) Linear rise time as a function of N . We fix field strength E_0 at 0.4 a.u., and thus $\Omega_R = E_0 d_{i,i+1} = 0.4$ a.u. Laser frequency is set to $\omega = \Delta_{\text{sg}} = 1$ a.u. Green open circles represent data points, and blue lines are linear fitting results for data points.

APPENDIX C: RISE TIME DEPENDING ON THE TOTAL NUMBER OF ENERGY LEVELS AND COUPLING STRENGTH IN THE EQUISPACED MODEL

We know that in the equispaced model, the cascade resonance occurs when the frequency condition is satisfied no matter how weak the electric field is. The electrons are excited to conduction levels in a cascade way from lower to higher. We can foresee that the weaker the field strength and the more energy levels there are, the longer it takes for the electrons to be excited to the highest energy level. Therefore, the rise time should depend on the total number of energy levels and the coupling strength. Figures 9(d) and 9(e) clearly show that the rise time exhibits a linear dependence of Ω_R^{-1} and N . We can describe it by

$$T_r \propto N/\Omega_R. \quad (\text{C1})$$

From the perspective of HHG, this relation indicates that the manipulation of the harmonic cutoff using laser duration is easier to achieve in a large-size nanoribbon material.

APPENDIX D: HHG IN GNRs BY APPLYING DRIVING FIELDS ALONG THE RIBBON DIRECTION

We study the harmonic generation in GNRs by applying the driver field to the ribbon direction. Figures 10(a) and 10(b), respectively, show the harmonic spectra for 40-AGNR and 40-ZGNR as a function of the field strength, and the wavelength is $3 \mu\text{m}$. It is found that the cutoffs of harmonic

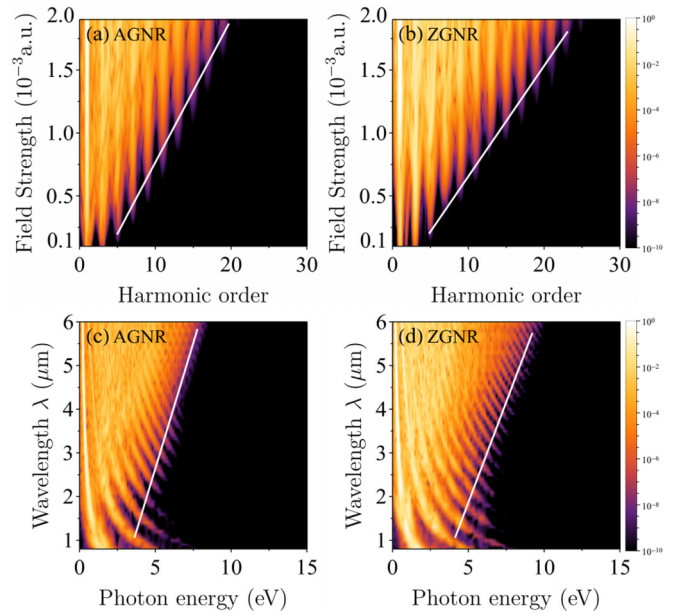


FIG. 10. (a),(b) Harmonic spectra vs field strength in 40-AGNR and 40-ZGNR, respectively, when the field is polarized along the ribbon direction. The laser frequency is fixed at $\omega_0 = 0.0152$ a.u. ($\lambda = 3 \mu\text{m}$). (c),(d) Harmonic spectra vs wavelength in 40-AGNR and 40-ZGNR, respectively. The field strength is $E_0 = 0.0012$ a.u. ($I_0 = 5.04 \times 10^{10} \text{ W/cm}^2$).

spectra for both AGNR and ZGNR scale linearly with the field strength; see the white lines. This is because the dispersion of subbands of GNRs near the Fermi level is almost linear with k_x . In Figs. 10(c) and 10(d), we also see the linear dependence between the cutoff energy and the laser wavelength, which

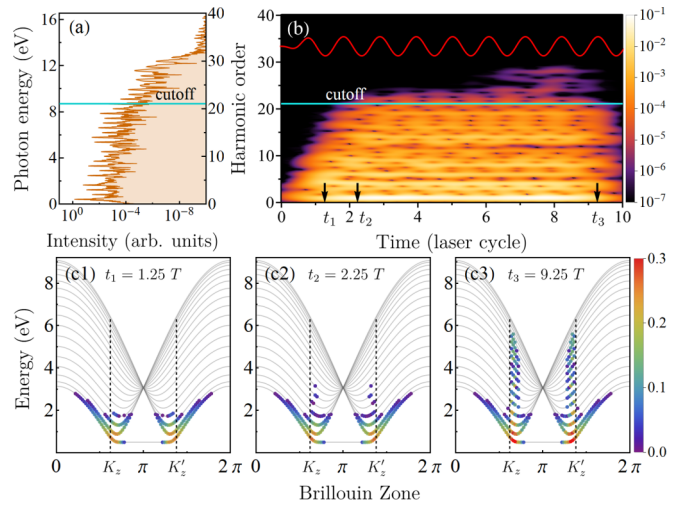


FIG. 11. (a) Harmonic spectrum for a 36-ZGNR with $E_g = 1.0$ eV. The field strength is $E_0 = 0.0012$ a.u. ($I_0 = 5.04 \times 10^{10} \text{ W/cm}^2$), and the field frequency matches a subband gap $\omega = 0.0152$ a.u. ($\lambda = 3 \mu\text{m}$). (b) Time-frequency distribution of the harmonic emission. The red curve is the vector potential of the laser field. The cyan line indicates the cutoff frequency. (c1)–(c3) Electron dynamics in the conduction subbands for the times t_i marked in (b).

agrees well with the general behavior of HHG in bulk solids. The reason is also attributed to the linear band dispersion.

APPENDIX E: HHG IN GNRs WITH ON-SITE POTENTIAL

In principle, we can open a fundamental gap in GNRs by adding staggered on-site potential on the two nearest-neighbor sites, respectively. Figure 11 shows the HHG spectrum and conduction dynamics in 36-ZGNR with a 1.0 eV gap. Because

there is a large fundamental gap between valence and conduction bands, the excitation rate decreases, and fewer electrons are excited to the first conduction band in Figs. 11(c1)–11(c3). However, one can still observe the cascade resonance, and the electrons still accumulate near the Dirac points, although the total population becomes lower in comparison to that of gapless 36-ZGNR. Correspondingly, the intensity of the HHG spectrum in Figs. 11(a) and 11(b) is lower than that in Figs. 3(a) and 11(b).

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