Beyond Planck-Einstein quanta: Amplitude-driven quantum excitation

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Linear-response quantum excitation is proportional to the amplitude of the field, with the energy of the excitation given by the driving frequency. As the amplitude is increased, there is a crossover, where the excitation energy is governed by the amplitude of the driving field, not its frequency. As the amplitude is increased even further, then complex quantum oscillations develop. We illustrate this phenomena with the exact solution of the simplest model of a charge-density-wave insulator driven by a spatially uniform time-dependent electric field. The driving by the field can be mapped onto a series of Landau-Zener problems, but with a complex, nonmonochromatic drive that varies for each momentum point in the reduced Brillouin zone.

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I. INTRODUCTION

In 1901, Planck introduced the idea of light quanta to calculate the spectrum for black body radiation [1], which was employed by Einstein in 1905 to explain the mysterious quantum properties of the photoelectric effect [2]. Later, the solution of the Landau-Zener tunneling problem in the 1930's [3,4], where a tunneling excitation is determined by the speed at which the minimal excitation gap is approached (and is proportional to the amplitude of an effective driving field), showed how the amplitude and not the frequency of a driving field can govern quantum excitation beyond linear response.

We consider one of the simplest problems in driven quantum systems: the excitation of an insulator across its gap due to applying a monochromatic or pulsed ac electric field. The energy transferred to the electrons by this driving frequency satisfies the Planck-Einstein relation [1,2] $E = \hbar\omega$ and is independent of the amplitude of the driving field, hence we expect to see no response until the frequency is large enough that $\hbar\omega \geqslant E_{\rm gap}$. Linear-response theory verifies this result, as the Kubo-Greenwood formula shows that the amplitude of the field just provides an overall scale to the response [5,6], and the ability to create an excitation is determined by energy conservation. As the amplitude of the field is increased, photons of a lower energy can combine together and create higher energy photons, and hence one would expect resonances at $\hbar\omega/2$ (or generally $\hbar\omega/n$ for multiphoton processes). However, the presence of a large field can also modify the quantum states themselves, and create states inside the gap region, thereby reducing the effective gap, and allowing excitations to occur at even lower frequencies.

The Landau-Zener tunneling problem has investigated some of these aspects [3,4]. While there is no applied field *per se* in this problem, one can assume that the rate at which the gap is approached is proportional to an effective driving field, and in this situation, it is known that the efficiency in tunneling to create excitations across the gap depends exponentially on the driving rate, and hence on the amplitude of the effective field. Indeed, the adiabatic theorem [7] guarantees no excitation for infinitesimally small fields with $\omega \to 0$. In addition, a generalized Landau-Zener problem, with the minimal crossing point passed numerous times due to an oscillatory drive, has been examined in the context of Landau-Zener interferometry [8–10]. These studies focus on the drive through the minimal crossing point occurring with a single frequency. In the work presented here, because the electric field couples in a nonlinear way to the electrons, the drive through the minimal crossing point is a highly nonlinear function of the driving field frequency, and hence it is not described by those results of Landau-Zener interferometry.

The Landau-Zener problem, and problems closely related to it, have been widely studied in many different contexts both in theory and in experiment. For example, in situations where the gap vanishes, one has the physical behavior of the Kibble-Zurek mechanism [11,12], which can be experimentally studied for linear driving, and for nonlinear (power-law) driving. In this context, a focus is placed on how the excitations are related to the critical exponents of the equilibrium system. We look at this problem in a different context. For us, there always is a gap, although the magnitude of the gap changes over the Brillouin zone, but the drive that is applied is complex, and varies from one momentum point to another, and can cross the minimal gap region multiple times. Summing over all momenta points corresponds to averaging over many Landau-Zener interferometry-like problems, one for each momentum (and with different gaps), each with an extremely nonmonochromatic driving field. Hence, this work is different from those studied elsewhere, and it typically has no single frequency that will govern the behavior of the system, nor does it generically satisfy scaling relations. These types of problems can be experimentally studied in many different systems, such as graphene [13].

The quantum excitation problem has also been examined in the field-induced ionization of atoms [14], in Josephson junctions [9], and in the photoelectric effect [15]. For the atomic problem, Keldysh [14] showed how a detailed quantum theory can describe the full regime from the frequency-driven excitation to the amplitude-driven excitation, and which has been applied recently to the photoelectric effect, where multiphoton excitations are observed when the photon light field has a large amplitude due to enhancement near an isolated sharp metallic tip [15]. But the quantum excitation problem in solid state systems is more complex than an ionization problem. As an electron is excited from a lower band to an

upper band, the state the particle has been excited to is blocked from further excitation. Such effects become quite important as the field becomes strong enough to excite significant fractions of electrons from the lower to the upper band. In addition, the oscillating current can both excite and deexcite electrons across the gap. These effects greatly complicate the net quantum excitation process.

Two questions immediately come to mind about field-driven quantum excitations in this context: (i) Does the excitation continue to depend exponentially on the amplitude so that nearly all of the excitation occurs near the maximal amplitude of a pulsed field, and (ii) as the amplitude is increased do we find a regime of purely amplitude-driven tunneling, where the excitation becomes independent of the driving frequency? We answer both of these questions with an exact solution for the nonlinear excitation of a solid. Note that this model has no interactions and does not thermalize. If the gap is large enough, the populations in the upper and lower bands will hardly change, even if interactions are added, because the thermalization will occur primarily within each subband.

In Sec. II, we present the formalism for the calculations, while results are presented in Sec. III. We provide a summary in Sec. IV.

II. FORMALISM

The charge-density-wave (CDW) model we use is that of spinless electrons moving on a lattice with a bipartite structure which has different site energies on the two sublattices (A/B). The Hamiltonian in the Schrödinger picture is

$$\mathcal{H}(t) = -\sum_{ij} \tau_{ij}(t) c_i^\dagger c_j + \sum_{i\in A}(U-\mu) c_i^\dagger c_i + \sum_{i\in B}(-\mu) c_i^\dagger c_i. \tag{1}$$

The first term is the kinetic energy, which involves a hopping between nearest neighbor lattice sites i and j with a hopping integral $\tau_{ij}(t)$ (the hopping matrix is a Hermitian matrix that is nonzero only for i and j nearest neighbors). The second and third terms include the chemical potential μ and the external potential U which is nonzero only on the A sublattice. We set $\mu = U/2$ in our calculations to have the case of half filling. Since the electrons do not interact with each other, the spin degree of freedom is trivial, and has been neglected here. The field is introduced via a time-dependent hopping integral, employing the Peierls substitution [16],

$$\tau_{ij}(t) = \frac{\tau^*}{2\sqrt{d}} \exp\left[-\frac{ie}{\hbar c}\int_{\mathbf{R}_i}^{\mathbf{R}_j} \mathbf{A}(t)\cdot d\mathbf{r}\right], \tag{2}$$

and we take the limit as $d \to \infty$ using τ^* as the energy unit. Here $\mathbf{A}(t)$ is the time-dependent (but spatially uniform) vector potential in the Hamiltonian gauge (where the scalar potential vanishes). The field is chosen to point in the diagonal direction $\mathbf{A}(t) = A_0(t)(1,1,1,\ldots)$ with $A_0(t)$ given by the antiderivative of the electric field as a function of time. In this case, the (time-dependent) electronic band structure becomes

$$\epsilon\left(\mathbf{k} - \frac{e\mathbf{A}(t)}{\hbar c}\right) = -\lim_{d\to\infty} \frac{\tau^*}{\sqrt{d}} \sum_{i=1}^{d} \cos\left[k_i - \frac{e\mathbf{A}_i(t)}{\hbar c}\right], \tag{3}$$

which is the standard form for the Peierls substitution [16]. Note that one of the advantages of this model is that the quantum excitation is driven solely by the Peierls substitution and requires no dipole matrix element that couples opposite parity bands. The field is treated semiclassically because it has a large amplitude. The Planck relation affects how the (quantum) electronic system responds to the (classical) drive.

For concreteness, we work in the infinite-dimensional limit, although the procedure produces an exact solution in any dimension (since the only effect of dimensionality is on the shape of the normal state density of states, we expect the infinite-dimensional results to be similar to those in two or higher dimensions; we choose this limit because it will make for easier comparison with dynamical mean-field theory calculations in the ordered phase). We take $\tau = \tau^*/2\sqrt{d}$ and set $\tau^* = 1$ as the energy unit. The band structure in the absence of a field has a gap of size $E_{\rm gap} = U$, with a density of states that is a mirror image on the A and B sublattices (see Fig. 1). There is a square-root-like singularity at the upper or lower band edge for the local density of states on each sublattice. The applied field is chosen to be either a monochromatic ac

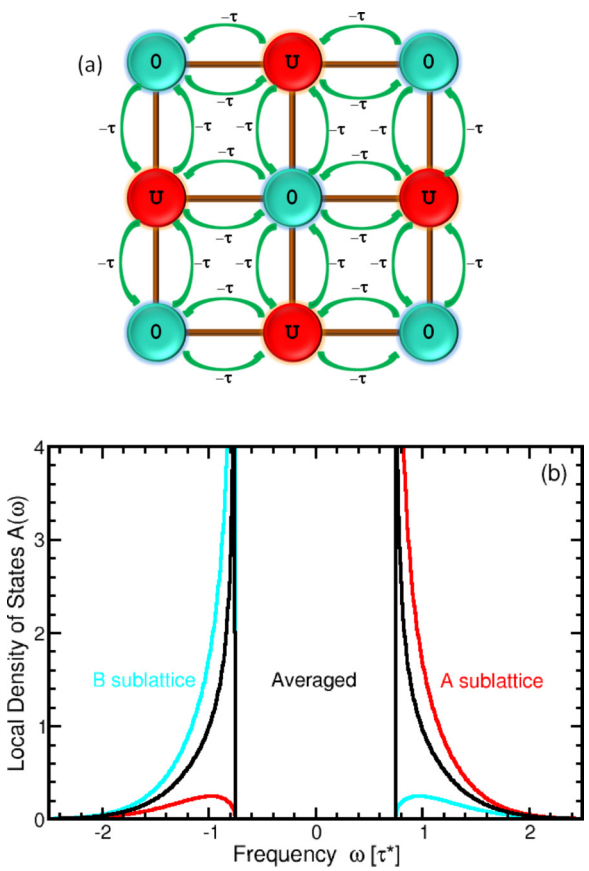

FIG. 1. (Color online) (a) Schematic of the charge-density-wave model on the checkerboard lattice. The A sublattice (red) has the on-site potential U, while the B sublattice (light blue) has a vanishing potential. Hopping is between nearest neighbors, as indicated by the green arrows. The schematic shows a two-dimensional square lattice, but we work on an infinite-dimensional hypercubic lattice. (b) Local density of states on the A/B sublattices (red/light blue) and the average density of states (black) in equilibrium for $U = 1.5$.

field turned on at time $t = 0$ or an oscillating pulse shape. The instantaneous spectra, under the application of this field, are independent of time, and hence there always exists a lower and upper band separated by a band gap of U with "eigenenergies" $E_{\pm}(k,t) = \pm\sqrt{\epsilon^2(k - eA_0(t)/\hbar c) + U^2/4}$. This can be seen by directly diagonalizing the Hamiltonian in the Schrödinger picture at any instant of time

$$\mathcal{H}_S(t) = \sum_k \overbrace{c_k^\dagger \quad c_{k+Q}^\dagger} \begin{pmatrix} \frac{U}{2} - \mu + \epsilon_k(t) & \frac{U}{2} \\ \frac{U}{2} & \frac{U}{2} - \mu - \epsilon_k(t) \end{pmatrix} \times \begin{pmatrix} c_k \\ c_{k+Q} \end{pmatrix} \tag{4}$$

in momentum space, where we have set $\epsilon_k(t) = \epsilon(k - eA_0(t)/\hbar c)$.

Since the field points in the diagonal direction, we can expand the time-dependent band structure to be

$$\epsilon_k(t) = \cos\left(\frac{eaA_0(t)}{\hbar c}\right)\epsilon_k + \sin\left(\frac{eaA_0(t)}{\hbar c}\right)\bar{\epsilon}_k, \tag{5}$$

which depends on both the band structure ϵ_k and the projection of the velocity along the direction of the field

$$\bar{\epsilon}_k = -\lim_{d\to\infty} \frac{t^*}{\sqrt{d}} \sum_{l=1}^{d} \sin(ak_l). \tag{6}$$

The problem can be solved exactly by employing the Kadanoff-Baym-Keldysh formalism for the contour-ordered Green's function and using the Trotter formula to evaluate the relevant 2×2 evolution operators for each coupled momenta $\mathbf{k}$ and $\mathbf{k} + \mathbf{Q}$ with $\mathbf{Q} = (\pi,\pi,\pi,\ldots)$, as described in detail in Ref. [17]. We sketch the critical formulas here.

To evaluate the Green's function, one first solves for the Heisenberg representation of the creation/annihilation operators, which yields

$$\begin{pmatrix} c_k(t) \\ c_{k+Q}(t) \end{pmatrix} = \mathcal{U}(k,t,t_0) \begin{pmatrix} c_k(t_0) \\ c_{k+Q}(t_0) \end{pmatrix}. \tag{7}$$

The time-evolution operator $\mathcal{U}(k,t,t')$ is a time ordered product for each momentum,

$$\mathcal{U}(k,t,t') = \mathcal{T}_t \exp\left[-i \int_{t'}^{t} d\bar{t} \begin{pmatrix} \frac{U}{2} - \mu + \epsilon_k(\bar{t}) & \frac{U}{2} \\ \frac{U}{2} & \frac{U}{2} - \mu - \epsilon_k(\bar{t}) \end{pmatrix} \right]. \tag{8}$$

Because of the time dependence, the Trotter formula is employed,

$$\mathcal{U}(k,t,t') = \mathcal{U}(k,t,t - \Delta t)\mathcal{U}(k,t - \Delta t,t - 2\Delta t)\cdots\mathcal{U}(k,t' + \Delta t,t'). \tag{9}$$

For a small time step Δt at time t, we have

$$\mathcal{U}(k,t,t - \Delta t) = \exp\left[-i\Delta t \begin{pmatrix} \frac{U}{2} - \mu + \epsilon_k(t - \Delta t/2) & \frac{U}{2} \\ \frac{U}{2} & \frac{U}{2} - \mu - \epsilon_k(t - \Delta t/2) \end{pmatrix} \right]. \tag{10}$$

This exponential can be exactly found since it is a 2×2 matrix, and we show the result for the case of interest of half filling, where $\mu = U/2$

$$\mathcal{U}(k,t,t - \Delta t) = \cos\left(\Delta t\sqrt{\epsilon_k^2\left(t - \frac{\Delta t}{2}\right) + \frac{U^2}{4}}\right)\mathbf{I} - i \begin{pmatrix} \epsilon_k\left(t - \frac{\Delta t}{2}\right) & \frac{U}{2} \\ \frac{U}{2} & -\epsilon_k\left(t - \frac{\Delta t}{2}\right) \end{pmatrix} \frac{\sin\left(\Delta t\sqrt{\epsilon_k^2\left(t - \frac{\Delta t}{2}\right) + \frac{U^2}{4}}\right)}{\sqrt{\epsilon_k^2\left(t - \frac{\Delta t}{2}\right) + \frac{U^2}{4}}}. \tag{11}$$

In our calculations, we start from a finite minimum time t_0, determining the evolution operator $\mathcal{U}(k,t,t_0)$ for all subsequent times. For each k, we find the two-time evolution operator from the identity

$$\mathcal{U}(k,t,t') = \mathcal{U}(k,t,t_0)\mathcal{U}^\dagger(k,t_0,t'). \tag{12}$$

Once the time evolution at each time pair is found, one can directly construct the retarded and lesser Green's functions as functions of momentum, or, after summing over momentum, determine the local Green's functions. We are particularly interested in the filling in the upper band, which requires one to use a gauge-invariant formalism to derive. The formulas are cumbersome, and appear in Appendix B of Ref. [17].

III. RESULTS

The electric field in each direction satisfies $E(t) = E_0 \sin(\omega_0 t)\theta(t)$ for the monochromatic ac field and $E(t) = E_0 \sin(\omega_0 t)\exp(-t^2/25)$ for the pulsed field. In Figs. 2(a) and 2(b), we examine the excitation process in the ac field with $\omega_0 = 1$ and $\omega_0 = 3$, respectively, for a CDW with a gap satisfying $U = 1.5$. Because an ac driving field produces a periodic response, we must determine the average occupancy of the upper instantaneous band in its "steady state," which is shown by the dashed lines just outside Fig. 2. Both systems approach this "steady state" relatively quickly, but in the lower-frequency case [Fig. 2(a)], the final density in the upper band does not have a monotonic dependence on the driving amplitude for this range of amplitudes of the field.

In Figs. 3(a) and 3(b), we show similar plots, but now for the pulsed case. Here we see quite different behavior. First off, a true steady state occurs, because at long times there is no field and the system dephases into a steady population in each band (transfer between bands can only occur when a field is on). Second, in the lower-frequency case [Fig. 3(a)], one can see the excitation is dominated by the regions where the field

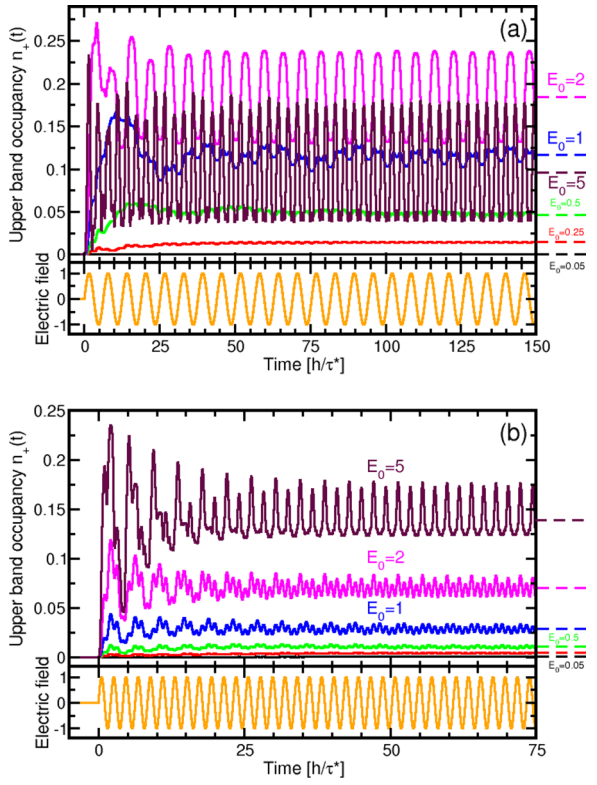

FIG. 2. (Color online) Time trace for the occupancy in the upper band as a function of time for an ac field driving of the CDW insulator with an equilibrium gap equal to $U = 1.5$ (started initially from $T = 0$) and two frequencies: (a) $\omega_0 = 1$ (less than the equilibrium gap) and (b) $\omega_0 = 3$ (larger than the equilibrium gap). We take as our measure of the final occupancy of the upper band the average value of the occupancy, averaged over one period in the long-time limit. These values are indicated by the dashed lines. The labels show the amplitude of the field. The red curve in (b) is for $E_0 = 0.25$. The field trace is plotted below each panel. Note that the time axis is half as long in (b).

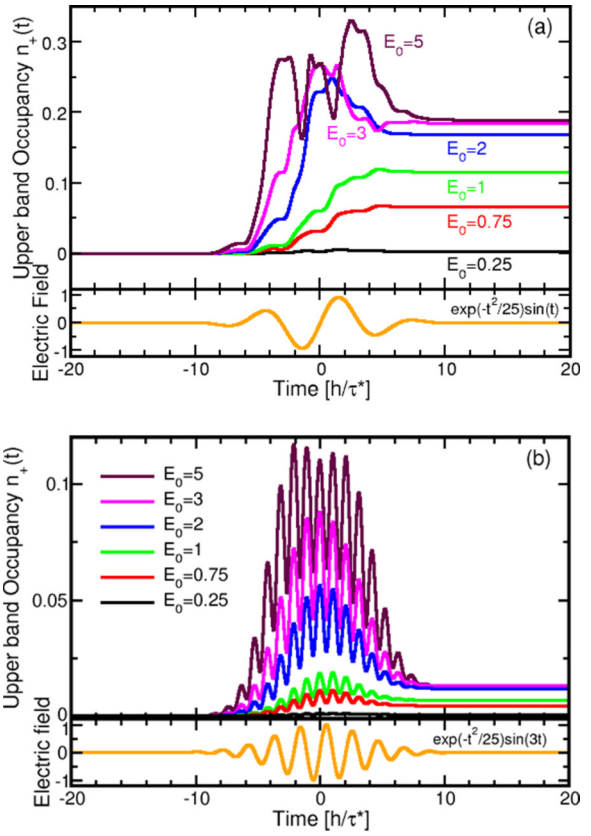

FIG. 3. (Color online) Excitation in the upper band as a function of time for the (a) low-frequency $\omega_0 = 1$ and (b) high-frequency $\omega_0 = 3$ pulsed-field cases. The labels in (a) and the legend in (b) show the amplitude of the field, which is plotted for the unit amplitude below each panel.

amplitude is the largest, but the full excitation occurs over an extended period of time, and certainly is not instantaneous. Third, the excitation is fairly monotonic in time, implying it is dominated by excitation processes and there is limited deexcitation. Note the steplike excitation for low-amplitude fields, which follow precisely the Landau-Zener picture of tunneling enhanced when the instantaneous magnitude of the field is large. In contrast, the higher-frequency case in Fig. 3(b) shows very dramatic deexcitation processes, and the final excitation requires one to examine the full time dependence of the system. It cannot be described just by the regions where the field amplitude is maximal. Instead, the quantum excitation is primarily determined by the lower field amplitudes near the start of the pulse, and the rest of the evolution corresponds to nearly equal excitation followed by deexcitation. In addition, none of the final distributions appear to be thermal, as expected, and are examined in further detail in Ref. [18].

We now examine spectroscopy for the long-time excitation curves as a function of the driving frequency for an ac drive (Fig. 4) and for the pulsed field (Fig. 5). In both cases, when the amplitude is small, the Planck-Einstein relation holds and no excitation occurs until the frequency becomes approximately equal to the gap (there is a small spread due to the finite spread of the Fourier transform of the applied electric field, especially for the pulsed case). As the amplitude is increased, we see the expected nonlinear effect of a peak forming at a frequency about one half of the gap size. But as the amplitude is increased further, the excitation spectra become quite flat in frequency, indicating the crossover to an amplitude-driven

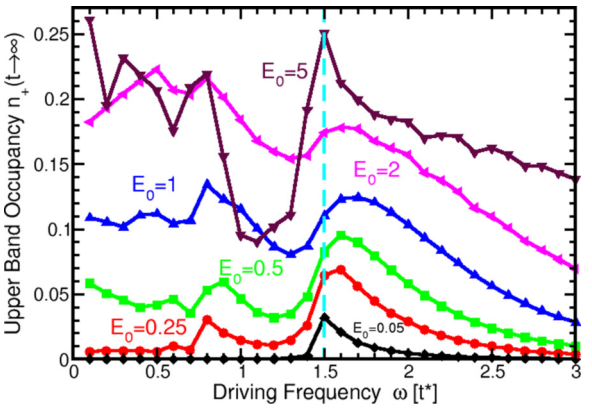

FIG. 4. (Color online) Upper band occupancy spectra for fixed amplitude and varying driving frequency with an ac field drive and the light blue dashed line showing where the equilibrium gap is.

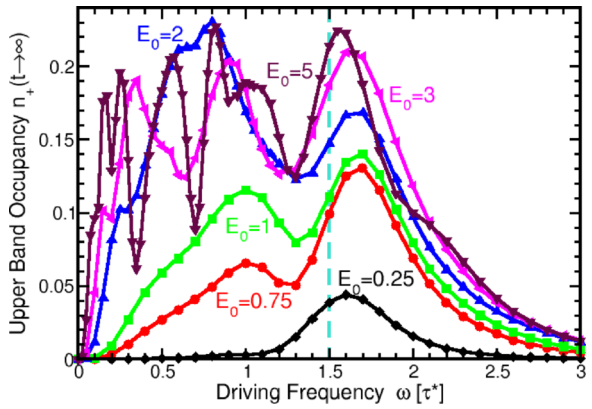

FIG. 5. (Color online) Excited state spectroscopy for the pulsed field, with the dashed blue line showing the value of the equilibrium gap. Here, the behavior is similar to Fig. 4, except the frequencies are less well defined because of the finite pulse width, which becomes particularly important in the low-frequency regime, and suppresses the signal there. The amplitude of each pulse is given by the color-coded labels near each respective curve.

excitation, although the curvature of the band structure never allows for a fully flat curve. Finally, as the amplitude is made even larger, we see interesting quantum oscillations develop in the spectra, which become more complex for larger amplitude driving. This behavior resembles the dynamical behavior shown by Shirley for two-level systems [19], and the appearance could be simply a result of the renormalization of the energies of multiphoton processes and a narrowing of the peaks with increasing amplitude. But it might also signal the onset of a new quantum regime that is governed primarily by the field amplitude and not the driving frequency (but with additional frequency-driven resonances at allowed multiphoton processes). This helps explain the universal dip of the spectra between half the gap and the gap, because no multiphoton processes are possible there.

We also can examine the scaling hypothesis of Refs. [9,10] which says we would expect $n_+(t \to \infty, E_0/\omega)$ to be a constant. If this holds, we expect a self-similar nature to the family of curves in Fig. 4. Comparing the results at $\omega = 1$ to $\omega = 3$ shows such a result approximately holds, but carefully comparing $\omega = 1$ to $\omega = 2$ shows it does not hold when the field amplitude becomes large. This is because of the averaging over many Landau-Zener transitions and the nonlinear nature of the Peierls substitution, which no longer allows the scaling relations to hold exactly.

IV. SUMMARY

In this paper, we have shown the crossover for how a quantum system is initially excited by the Planck-Einstein quanta, but then nonlinear effects change the behavior first into excitations arising from the nonlinear combination of photons to create high energy excitations until the excitation is dominated by the amplitude of the driving field and shows limited frequency dependence. At the largest amplitudes, additional quantum oscillations occur, whose origins are likely due to different high-field quantum effects. This paper shows a way to examine the details behind quantum excitation in strong fields that goes beyond the common belief that tunneling phenomena are dominated by the region in time where the field amplitude is the largest. We clearly see more complex and rich phenomena arising in this limit. Initial experiments that have examined this phenomena have been performed in cold atom systems on tunable hexagonal lattices [20].

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