

Higher-harmonic generation in the driven Mott-Hubbard model

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Using Floquet theory and the hierarchy of correlations, we study the non-equilibrium dynamics of the Mott insulator state in the Fermi-Hubbard model under the influence of a harmonically oscillating electric field representing the pump laser. After deriving the associated Floquet exponents, we consider higher-harmonic generation where the strongest signal is obtained if the driving frequency equals one third of the Mott gap.

I. INTRODUCTION

Non-equilibrium dynamics of interacting quantum many-body systems are a rich and complex field displaying many fascinating phenomena – especially for strongly interacting systems. As the *drosophila* of strongly interacting quantum many-body systems, we consider the Fermi-Hubbard model. The non-equilibrium dynamics can be generated by an external stimulus, such as a pump laser [1–4], which we model as a harmonically oscillating electric field – facilitating a Floquet analysis [5, 6].

Even for a static electric field, the Fermi-Hubbard model displays interesting phenomena [13–16], such as doublon-holon pair creation (dielectric breakdown, see, e.g., [7–12]) which can also be understood in a Floquet picture. For harmonically oscillating fields, one can employ Floquet engineering [17, 18], for example in order to suppress dissipation [19] or to modify pre-thermalization dynamics [20, 21]. In the following, we shall study higher-harmonic generation in the harmonically driven Fermi-Hubbard model [22–25]. To this end, we combine Floquet theory with the method of the hierarchy of correlations [26]. Note that, in contrast to many other approaches, we do not apply the Floquet expansion directly to the quantum state [27], but to the time-dependent correlation functions.

II. DRIVEN FERMION-HUBBARD MODEL

To analyze the dynamics of a prototypical strongly interacting quantum many-body system under periodic driving, we consider the fermionic Hubbard model coupled to an external electric field. The Hamiltonian governing the system is given by ($\hbar = 1$)

$$\hat{H}_{\text{ext}} = -\frac{1}{Z} \sum_{\mu\nu s} T_{\mu\nu} \hat{c}_{\mu s}^\dagger \hat{c}_{\nu s} + U \sum_{\mu} \hat{n}_{\mu}^{\uparrow} \hat{n}_{\mu}^{\downarrow} + \sum_{\mu s} V_{\mu}(t) \hat{n}_{\mu s}. \quad (1)$$

Here, $V_{\mu}(t) = q\mathbf{x}_{\mu} \cdot \mathbf{E}(t)$ represents the time-dependent potential energy of an electron with charge q at the posi-

tion \mathbf{x}_{μ} of site μ , where $\mathbf{E}(t)$ is the external electric field. Assuming that the wavelength of the pump laser is much larger than all other relevant length scales, we neglect its spatial dependence and approximate it by a purely time-dependent external electric field $\mathbf{E}(t)$.

The fermionic creation and annihilation operators at sites μ and ν , with spin $s \in \uparrow, \downarrow$, are denoted as $\hat{c}_{\mu s}^\dagger$ and $\hat{c}_{\nu s}$, respectively. The hopping matrix $T_{\mu\nu}$ takes the value of the tunneling rate T for nearest neighbors, and zero otherwise. The coordination number Z represents the number of nearest neighbors for a given lattice site.

To account for the potential term, we perform a Peierls substitution, leading to complex-valued entries for the hopping matrix. The time-dependent hopping matrix is then expressed as

$$T_{\mu\nu}(t) = T_{\mu\nu} \exp \left\{ -i \int_0^t dt' [V_{\mu}(t') - V_{\nu}(t')] \right\}. \quad (2)$$

In this way, we obtain the explicitly time-dependent Hamiltonian

$$\hat{H}(t) = -\frac{1}{Z} \sum_{\mu\nu s} T_{\mu\nu}(t) \hat{c}_{\mu s}^\dagger \hat{c}_{\nu s} + U \sum_{\mu} \hat{n}_{\mu}^{\uparrow} \hat{n}_{\mu}^{\downarrow}. \quad (3)$$

Similar to a gauge transformation in electrodynamics, the scalar potential $V_{\mu}(t)$ is transferred to the hopping matrix $T_{\mu\nu}(t)$, whose phase is analogous to that given by the vector potential, while the on-site repulsion term U remains unchanged.

A. The hierarchy of correlations

To obtain approximate solutions, we utilize a hierarchical method suitable for systems with large coordination numbers Z . To do this, we separate the reduced density matrices into correlations between lattice sites and on-site density matrices. For example, the correlation between two sites is given by $\hat{\rho}_{\mu\nu}^{\text{corr}} = \hat{\rho}_{\mu\nu} - \hat{\rho}_{\mu} \hat{\rho}_{\nu}$. Likewise, correlations among three sites can be expressed as $\hat{\rho}_{\mu\nu\lambda}^{\text{corr}} = \hat{\rho}_{\mu\nu\lambda} - \hat{\rho}_{\mu} \hat{\rho}_{\nu} \hat{\rho}_{\lambda} - \hat{\rho}_{\mu\nu}^{\text{corr}} \hat{\rho}_{\lambda} - \hat{\rho}_{\mu\lambda}^{\text{corr}} \hat{\rho}_{\nu} - \hat{\rho}_{\nu\lambda}^{\text{corr}} \hat{\rho}_{\mu}$, etc.

The on-site density matrix $\hat{\rho}_{\mu}$ and the two-site correlators $\hat{\rho}_{\mu\nu}^{\text{corr}}$ obey evolution equations, which can be

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schematically represented as follows [26]

$$i\partial_t \hat{\rho}_\mu = F_1(\hat{\rho}_\mu, \hat{\rho}_{\mu\nu}^{\text{corr}}) = \mathcal{O}(1) \quad (4)$$

$$i\partial_t \hat{\rho}_{\mu\nu}^{\text{corr}} = F_2(\hat{\rho}_\mu, \hat{\rho}_{\mu\nu}^{\text{corr}}, \hat{\rho}_{\mu\nu\lambda}^{\text{corr}}) = \mathcal{O}(1/Z). \quad (5)$$

Similar equations hold for higher-order correlations. The specific forms of the nonlinear functions F_n are determined by the exact von Neumann equation for the density matrix of the Hubbard model. Analyzing the evolution equations for the correlators reveals [28, 29] that the scaling hierarchy remains preserved over time when the initial values satisfy $\hat{\rho}_\mu = \mathcal{O}(1)$, $\hat{\rho}_{\mu\nu}^{\text{corr}} = \mathcal{O}(1/Z)$, etc.

As the two-site correlators are of order $1/Z$, we can approximate Equation (4) as $i\partial_t \hat{\rho}_\mu \approx F_1(\hat{\rho}_\mu, 0)$. The zeroth-order solution $\hat{\rho}_\mu^0$ determines the mean field background of the system and will be discussed in the subsequent section. Similarly, since the three-point correlators scale with $1/Z^2$, we can approximate Equation (5) as $i\partial_t \hat{\rho}_{\mu\nu}^{\text{corr}} \approx F_2(\hat{\rho}_\mu, \hat{\rho}_{\mu\nu}^{\text{corr}}, 0)$. Introducing the following operators proves to be advantageous when analyzing the evolution equation

$$\hat{c}_{\mu s I} = \hat{c}_{\mu s} \hat{n}_{\mu \bar{s}}^I = \begin{cases} \hat{c}_{\mu s} (1 - \hat{n}_{\mu \bar{s}}) & \text{for } I = 0 \\ \hat{c}_{\mu s} \hat{n}_{\mu \bar{s}} & \text{for } I = 1 \end{cases}. \quad (6)$$

Here \bar{s} denotes the opposite spin to s . The dynamics of the two-site correlation functions to first order $\mathcal{O}(1/Z)$ takes then the form

$$\begin{aligned} i\partial_t \langle \hat{c}_{\mu s I}^\dagger \hat{c}_{\nu s J} \rangle^{\text{corr}} &= \frac{1}{Z} \sum_{\lambda L} T_{\mu\lambda}(t) \langle \hat{n}_{\mu \bar{s}}^I \rangle^0 \langle \hat{c}_{\lambda s L}^\dagger \hat{c}_{\nu s J} \rangle^{\text{corr}} \\ &\quad - \frac{1}{Z} \sum_{\lambda L} T_{\lambda\nu}(t) \langle \hat{n}_{\nu \bar{s}}^J \rangle^0 \langle \hat{c}_{\mu s I}^\dagger \hat{c}_{\lambda s L} \rangle^{\text{corr}} \\ &\quad + U(J - I) \langle \hat{c}_{\mu s I}^\dagger \hat{c}_{\nu s J} \rangle^{\text{corr}} \\ &\quad + \frac{T_{\mu\nu}(t)}{Z} (\langle \hat{n}_{\mu \bar{s}}^I \rangle^0 \langle \hat{n}_{\nu s}^1 \hat{n}_{\nu \bar{s}}^J \rangle^0 - \langle \hat{n}_{\nu \bar{s}}^J \rangle^0 \langle \hat{n}_{\mu s}^1 \hat{n}_{\mu \bar{s}}^I \rangle^0), \end{aligned} \quad (7)$$

where the expectation values $\langle \hat{X}_\mu \rangle^0$ are taken with the zeroth-order solution $\langle \hat{X}_\mu \rangle^0 = \text{Tr}\{\hat{X}_\mu \hat{\rho}_\mu^0\}$. For a given mean-field background $\hat{\rho}_\mu^0$, these differential equations describe the free dynamics of the quasi-particle excitations. In the absence of an electric field, Equation (7) yields the dispersion of the quasi-particle excitations.

While one approach to studying the equations of motion involves factorizing them [30], leading to an effective Dirac equation for the quasi-particle operators [31], in this case, we opt for a direct analysis focusing on the dynamics of the correlations.

III. FLOQUET ANALYSIS

In order to solve Equation (7), we have so specify the mean-field background $\hat{\rho}_\mu^0$. In the strong-coupling regime $U \gg T$, the Mott insulator state has one particle per site (neglecting virtual hopping corrections with probabilities

of order T^2/U^2), which merely leaves the spin structure to be determined.

In the strong-coupling limit $U \rightarrow \infty$, any finite temperature would be below U (i.e., the Mott gap), but above T^2/U (i.e., the energy scale of spin fluctuations). Thus, such a temperature is sufficiently low to prevent the generation of doublon-holon pairs, but it would have the tendency to disrupt the spin order within the system. Consequently, we choose the following ansatz for a spin-disordered background

$$\hat{\rho}_\mu \approx \hat{\rho}_\mu^0 = \frac{|\uparrow\rangle_\mu \langle \uparrow| + |\downarrow\rangle_\mu \langle \downarrow|}{2}. \quad (8)$$

By using Equation (4), it can be verified that this state remains inert to lowest order $\mathcal{O}(1)$. Only when considering the back-reaction of the correlators on the mean field, an amplitude for the generation of doublon-holon pairs arises at next order $\mathcal{O}(1/Z)$.

Within the leading order of the hierarchical expansion, the modes of the quasi-particle excitations decouple. Specifically, after performing a Fourier transform of Equation (7), we observe that each mode evolves according to

$$\begin{aligned} i\partial_t f_{\mathbf{k}s}^{IJ} &= (J - I)U f_{\mathbf{k}s}^{IJ} \\ &\quad + \frac{T_{\mathbf{k}}(t)}{2} \sum_L (f_{\mathbf{k}s}^{LJ} - f_{\mathbf{k}s}^{IL}) + (I - J) \frac{T_{\mathbf{k}}(t)}{4}. \end{aligned} \quad (9)$$

As explained above, the electrical field is encoded in the Fourier components of the hopping matrix, which adopts the usual minimal coupling form $T_{\mathbf{k}}(t) = T_{\mathbf{k}-q\mathbf{A}(t)}$ with $\partial_t \mathbf{A}(t) = -\mathbf{E}(t)$. For simplicity, we assume a periodic driving field of the form $\mathbf{E} = \mathbf{E}_0 \cos(\omega t)$, i.e., in linear polarization, where ω is the driving frequency.

As the next step, we Fourier decompose the time dependence of $T_{\mathbf{k}}(t)$

$$T_{\mathbf{k}}(t) = T_{\mathbf{k}}^\perp + \sum_n T_{\mathbf{k},n}^\parallel e^{-in\omega t}, \quad (10)$$

where $T_{\mathbf{k}}^\perp$ contains the contribution perpendicular to the electric field (which is thus independent of time) while the $T_{\mathbf{k},n}^\parallel$ encode the time-dependence induced by the electric field (i.e., the Floquet channels).

E.g., for a square lattice with the lattice spacing ℓ , where the electric field \mathbf{E}_0 is aligned with one of the principal axes, these expansion coefficients $T_{\mathbf{k},n}^\parallel$ can be expressed in terms of Bessel functions of the first kind. For even values of n , the coefficients are real, taking the form $T_{\mathbf{k},n}^\parallel = (2T/Z) \cos(k^\parallel \ell) \mathcal{J}_n(qE_0 \ell / \omega)$. On the other hand, for odd values of n , the coefficients are purely imaginary, given by $T_{\mathbf{k},n}^\parallel = -i(2T/Z) \sin(k^\parallel \ell) \mathcal{J}_n(qE_0 \ell / \omega)$.

Applying Floquet theory, we solve the set of equations (9) via an analogous expansion (10) into Floquet channels. To this end, let us combine the four components $f_{\mathbf{k}s}^{IJ}$ into one vector $\mathcal{F}_{\mathbf{k}s}$ and write (9) as

$$i\partial_t \mathcal{F}_{\mathbf{k}s}(t) = \mathcal{M}_{\mathbf{k}}(t) \cdot \mathcal{F}_{\mathbf{k}s}(t) + \mathcal{S}_{\mathbf{k}}(t), \quad (11)$$

with 4×4 matrices $\mathcal{M}_{\mathbf{k}}(t)$. The source terms $\mathcal{S}_{\mathbf{k}}(t)$ can be eliminated by a simple shift of the $\mathcal{F}_{\mathbf{k}s}$. Now we may expand $\mathcal{M}_{\mathbf{k}}(t)$ and $\mathcal{F}_{\mathbf{k}s}(t)$ into Floquet channels as in (10) and arrive at

$$i\partial_t \tilde{\mathfrak{F}}_{\mathbf{k}s}(t) = \mathfrak{M}_{\mathbf{k}} \cdot \tilde{\mathfrak{F}}_{\mathbf{k}s}(t), \quad (12)$$

where $\tilde{\mathfrak{F}}_{\mathbf{k}s}(t)$ is now an infinite dimensional vector containing all the Floquet channels $n \in \mathbb{Z}$. In analogy, the matrix $\mathfrak{M}_{\mathbf{k}}$ is infinite dimensional as it contains the coupling between all the Floquet channels – but it is independent of time. The eigenvectors of the matrix $\mathfrak{M}_{\mathbf{k}}$ correspond to the Floquet states whereas the associated eigenvalues $\omega\nu_{\mathbf{k}}$ yield the Floquet exponents $\nu_{\mathbf{k}}$.

Translating the above analysis back to our problem suggests the following ansatz for the correlators

$$f_{\mathbf{k},s}^{01} = \sum_n F_{\mathbf{k},n}^{01} e^{i(n+\nu_{\mathbf{k}})\omega t}, \quad (13)$$

$$f_{\mathbf{k},s}^{00} + \frac{1}{4} = \sum_n G_{\mathbf{k},n} e^{i(n+\nu_{\mathbf{k}})\omega t} = -f_{\mathbf{k},s}^{11} + \frac{1}{4}, \quad (14)$$

$$f_{\mathbf{k},s}^{10} = \sum_n F_{\mathbf{k},n}^{10} e^{i(n+\nu_{\mathbf{k}})\omega t}. \quad (15)$$

Here, we exploited the particle-hole symmetry $f_{\mathbf{k},s}^{00} = -f_{\mathbf{k},s}^{11}$ which is a direct consequence of equations (9).

For the eigenvalue equation

$$\mathfrak{M}_{\mathbf{k}} \cdot \tilde{\mathfrak{F}}_{\mathbf{k}s}^{\nu_{\mathbf{k}}} = \omega\nu_{\mathbf{k}} \tilde{\mathfrak{F}}_{\mathbf{k}s}^{\nu_{\mathbf{k}}}, \quad (16)$$

it can be shown that the $\nu_{\mathbf{k}}$ values are real, and the solutions always occur in pairs. Specifically, for each solution with Floquet exponent $\nu_{\mathbf{k}}$ and eigenvector $\tilde{\mathfrak{F}}_{\mathbf{k}s}^{\nu_{\mathbf{k}}}$ containing the expansion coefficients $F_{\mathbf{k},n}^{01}$, $F_{\mathbf{k},n}^{10}$, and $G_{\mathbf{k},n}$, there exists a conjugate solution with the opposite Floquet exponent $\tilde{\nu}_{\mathbf{k}} = -\nu_{\mathbf{k}}$. Its eigenvector $\tilde{\tilde{\mathfrak{F}}}_{\mathbf{k}s}^{\nu_{\mathbf{k}}}$ contains the entries

$$\tilde{F}_{\mathbf{k},n}^{01} = (F_{\mathbf{k},-n}^{10})^*, \quad \tilde{F}_{\mathbf{k},n}^{10} = (F_{\mathbf{k},-n}^{01})^*, \quad \tilde{G}_{\mathbf{k},n} = (G_{\mathbf{k},-n})^*. \quad (17)$$

Furthermore, from $[\langle c_{\mu s I}^\dagger c_{\nu s J} \rangle^{\text{corr}}]^* = \langle c_{\nu s J}^\dagger c_{\mu s I} \rangle^{\text{corr}}$, we deduce the property $f_{\mathbf{k},s}^{IJ} = (f_{\mathbf{k},s}^{JI})^*$. As a result, the Floquet correlators are linear combinations of the conjugate solutions

$$\begin{aligned} f_{\mathbf{k},s}^{01} &= \sum_n \left(F_{\mathbf{k},n}^{01} e^{i(n+\nu_{\mathbf{k}})\omega t} + (F_{\mathbf{k},n}^{10})^* e^{-i(n+\nu_{\mathbf{k}})\omega t} \right) \\ &= (f_{\mathbf{k},s}^{10})^* \end{aligned} \quad (18)$$

and

$$\begin{aligned} f_{\mathbf{k},s}^{00} + \frac{1}{4} &= \sum_n \left(G_{\mathbf{k},n} e^{i(n+\nu_{\mathbf{k}})\omega t} + (G_{\mathbf{k},n})^* e^{-i(n+\nu_{\mathbf{k}})\omega t} \right) \\ &= -f_{\mathbf{k},s}^{11} + \frac{1}{4}. \end{aligned} \quad (19)$$

A remaining issue concerns the proper normalization of the correlators. Since we applied the mode expansion to correlation functions instead of a state vector of the system, the eigenvectors $\tilde{\mathfrak{F}}_{\mathbf{k}s}^{\nu_{\mathbf{k}}}$ are not normalized to unity. Therefore, an additional condition is required to specify the appropriate normalization of the correlators.

In the case of the mean-field background (8), we observe that the evolution equations (9) leave the quantity $f_{\mathbf{k},s}^{01} f_{\mathbf{k},s}^{10} + (f_{\mathbf{k},s}^{00} + 1/4)^2 = 1/16$ invariant. From this condition, we deduce that in leading order of the hierarchy, the quasi-particle modes remain bounded regardless of the form of the applied electrical field. Furthermore, we can derive the normalization condition

$$\sum_n \left[|F_{\mathbf{k},n}^{01}|^2 + |F_{\mathbf{k},n}^{10}|^2 + 2|G_{\mathbf{k},n}^{00}|^2 \right] = \frac{1}{16}, \quad (20)$$

where we have employed the Fourier expansions (18) and (19). It is important to note that this condition explicitly differs from the usual vector norm of $\tilde{\mathfrak{F}}_{\mathbf{k}s}^{\nu_{\mathbf{k}}}$.

A. Floquet exponents

The eigenvalue equation (16) has solutions for both integer and non-integer values of $\nu_{\mathbf{k}}$. Furthermore, given any solution $\nu_{\mathbf{k}}$, one can construct other solutions with $\nu_{\mathbf{k}} \pm 1$ by a simple transformation of the eigenvectors $\tilde{\mathfrak{F}}_{\mathbf{k}s}^{\nu_{\mathbf{k}}}$, see Appendix A. If we now focus on the non-integer Floquet exponents near zero, i.e., $0 < \nu_{\mathbf{k}} \ll 1$, they are related to the characteristic time scale $\tau_{\mathbf{k}} \sim 1/(\nu_{\mathbf{k}}\omega)$ of the growth of the correlation functions, for example near a resonant frequency $\omega = U/n$.

For small hopping, specifically $T/\omega \ll 1$ and $T/U \ll 1$, it is possible to determine these Floquet exponents from the eigenvalue equation (16). After some algebra (see Appendix A), we obtain the following expression [32]:

$$\sin^2(\pi\nu_{\mathbf{k}}) = \sin^2\left(\frac{\pi U}{\omega}\right) + \sin\left[\frac{2\pi U}{\omega}\right] \frac{\pi}{2\omega} \left(\frac{(T_{\mathbf{k}}^\perp)^2}{U} + \frac{2T_{\mathbf{k}}^\perp T_{\mathbf{k},0}^\parallel}{U} + \sum_n |T_{\mathbf{k},n}^\parallel|^2 \frac{U}{U^2 - \omega^2 n^2} \right) + \mathcal{O}(T^4). \quad (21)$$

Note that the apparent singularity from the denominator

$U^2 - \omega^2 n^2$ at resonance $\omega = U/n$ is compensated by the

pre-factor $\sin(2\pi U/\omega)$ which also vanishes at $\omega = U/n$.

When the driving frequency approaches the resonance condition $\omega = U/n$, the above relation yields a simple scaling law for small electric fields $qE_0\ell \ll U$

$$\nu_{\mathbf{k}} \approx \frac{\sqrt{2}nT}{ZU} \frac{1}{2^{2n}} \left(\frac{qE_0\ell n}{U} \right)^n \begin{cases} \cos(k^\parallel \ell) & \text{for } n \text{ even} \\ \sin(k^\parallel \ell) & \text{for } n \text{ odd,} \end{cases} \quad (22)$$

where we have inserted the results for the square lattice as a specific example.

More generally, near the n -th resonance, the Floquet exponent scales with E_0^n . This is consistent with the intuitive picture that absorbing n photons is necessary to overcome the energy gap U of the system. Since the process of multi-photon absorption (for weak fields E_0) should occur on a timescale that increases with the number of photons involved, we expect a slower increase around the higher-order resonances. Indeed, the characteristic timescale in which the correlations build up scales with powers of the electrical field, specifically $\tau_{\mathbf{k}} \sim (\omega/E_0)^n (Z/T)$.

IV. HIGHER HARMONICS

After having developed the Floquet analysis of the doublon-holon correlators in the driven Fermi-Hubbard mode, we may now turn to specific applications.

For example, one might envision switching on the pump laser suddenly, which corresponds to a quantum quench. After preparing the Hubbard system initially in the ground state (with respect to the doublon-holon quasi-particle excitations, not the spin structure), the system is driven into an excited state far from equilibrium by this sudden switching. Within our level of approximation, this initial state is then translated into a superposition of eigenstates of the Floquet system (16) for each mode \mathbf{k}, s . The various eigenvalues $\nu_{\mathbf{k}}$ occurring in this superposition would then determine the individual oscillation or initial growth of these Floquet states.

In the following, we consider a slow switching of the pump laser instead – which is probably closer to a typical experimental situation. Again starting in the ground state, the system is stationary for $E_0 = 0$, and thus the left-hand side of (9) will vanish. As a result, the system will be in a state with zero eigenvalue $\nu_{\mathbf{k}} = 0$. If the pump laser is now turned on adiabatically – as described by a matrix $\mathfrak{M}_{\mathbf{k}}(t)$ with a very slow time-dependence – the system will remain in the Floquet state corresponding to $\nu_{\mathbf{k}} = 0$. This, together with the normalization condition (20), allows us to evaluate the correlators $F_{\mathbf{k},n}^{01}$, $F_{\mathbf{k},n}^{10}$, and $G_{\mathbf{k},n}$.

To relate our findings to observable quantities, we consider the particle current between neighboring lattice sites μ and ν (for which $T_{\mu\nu} = T$)

$$J = i \sum_s \langle \hat{c}_{\mu,s}^\dagger \hat{c}_{\nu,s} \rangle + \text{h.c.} \quad (23)$$

In general, this current could also have a component perpendicular to the pump field \mathbf{E}_0 , but for our example of the square lattice, it is purely parallel and reads

$$J = -4 \int_{\mathbf{k}} \sin(k^\parallel \ell) (f_{\mathbf{k},s}^{01} + f_{\mathbf{k},s}^{10}). \quad (24)$$

Due to the anti-symmetry of the integrand in (24) with respect to k^\parallel and the symmetry properties of the $T_{\mathbf{k},n}^\parallel$, only odd multiples of higher harmonics occur, and we have $J(t) = \sum_n J_n \sin(n\omega t)$, where $n = 1, 3, 5, \dots$

Close to a resonance at $\omega n = U$, the current is dominated by a single correlator

$$F_{\mathbf{k},n}^{10} \approx - \frac{UT_{\mathbf{k},n}^\parallel}{2\sqrt{T_{\mathbf{k}}^4 + 4U^2 |T_{\mathbf{k},n}^\parallel|^2}}, \quad (25)$$

while all other correlators are suppressed by at least a factor of T/U . Evaluating the integration over the Brillouin zone for small electrical fields ($qE_0\ell/U \ll 1$), we obtain the resonant current as

$$J(t) \approx 1.4 \sqrt{\frac{U}{2T} \left(\frac{nqE_0\ell}{U} \right)^n} \frac{1}{2^{2n}} \sin(Ut). \quad (26)$$

The non-polynomial dependence of the current on the involved quantities U , T and E_0 indicates that it should be non-trivial to obtain this result via some sort of perturbation theory. Time-dependent perturbation theory could be used to derive the initial growth of correlations after suddenly turning on the pump laser (as described above), but here we are considering a steady state.

For the fundamental resonance $\omega = U$, the current scales with $\sqrt{qE_0\ell/U}$. This current would then emit electromagnetic radiation with the same frequency $\omega = U$ as the incident light. In order to emit higher frequencies, one could use the third harmonic $\omega = U/3$ where the current is suppressed by an additional factor of $qE_0\ell/U$.

V. CONCLUSIONS AND OUTLOOK

By combining Floquet theory with the method of the hierarchy of correlations, we study the non-equilibrium dynamics of the driven Fermi-Hubbard model under the influence of a harmonically oscillating electric field, which serves as a model for the pump laser. We derive the associated Floquet exponents, which determine the time scales of the evolution, such as the initial growth of the correlations after suddenly switching on the pump laser.

After developing the basic theory, we study higher-harmonic generation in a steady state, which can be reached by gradually turning on the pump laser. To this end, we calculate the time-dependent current which can be obtained from the correlations (or coherences) between neighboring sites. The ability to calculate such a quantity involving operators at two lattice sites highlights

one of the advantages of the method of the hierarchy of correlations in comparison to other approaches which effectively map the system onto a single lattice site (such as dynamical mean-field theory DMFT [33–39]).

As an outlook, our results can be used as an ingredient to study the energy balance of the steady Floquet states, where the energy absorption from the pump laser is compensated by the emission (including the higher-harmonics). Going from quasi-stationary Floquet states to more general non-stationary states (i.e., superpositions of Floquet states), the formalism developed above can also be used to investigate pre-thermalization [20, 40–44] and other phenomena.

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Appendix A: Floquet eigensystem

Utilizing the Floquet expansions (13) - (15) for the correlators, the system of algebraic equations adopts the following structure

$$-\omega(n + \nu_{\mathbf{k}})G_{\mathbf{k},n}(\nu_{\mathbf{k}}) - \frac{T_{\mathbf{k}}^{\perp}}{2}[F_{\mathbf{k},n}^{10}(\nu_{\mathbf{k}}) - F_{\mathbf{k},n}^{01}(\nu_{\mathbf{k}})] \\ = \frac{1}{2} \sum_m T_{\mathbf{k},m}^{\parallel} [F_{\mathbf{k},n+m}^{10}(\nu_{\mathbf{k}}) - F_{\mathbf{k},n+m}^{01}(\nu_{\mathbf{k}})] \quad (\text{A1})$$

$$[-\omega(n + \nu_{\mathbf{k}}) - U] F_{\mathbf{k},n}^{01}(\nu_{\mathbf{k}}) + T_{\mathbf{k}}^{\perp} G_{\mathbf{k},n}(\nu_{\mathbf{k}}) \\ = - \sum_m T_{\mathbf{k},m}^{\parallel} G_{\mathbf{k},n+m}(\nu_{\mathbf{k}}) \quad (\text{A2})$$

$$[-\omega(n + \nu_{\mathbf{k}}) + U] F_{\mathbf{k},n}^{10}(\nu_{\mathbf{k}}) - T_{\mathbf{k}}^{\perp} G_{\mathbf{k},n}^{00}(\nu_{\mathbf{k}}) \\ = \sum_m T_{\mathbf{k},m}^{\parallel} G_{\mathbf{k},n+m}(\nu_{\mathbf{k}}) \quad (\text{A3})$$

Here, we have explicitly denoted that the coefficients belong to the eigenvector with eigenvalue $\nu_{\mathbf{k}}$.

If our objective is to derive the eigenvector for the eigenvalue $\nu_{\mathbf{k}} + l$, where l is a natural number, we can achieve this by performing an index shift $n \rightarrow n - l$, resulting in the following correspondences

$$F_{\mathbf{k},n}^{01}(\nu_{\mathbf{k}} + l) = F_{\mathbf{k},n+l}^{01}(\nu_{\mathbf{k}}) \quad (\text{A4})$$

$$F_{\mathbf{k},n}^{10}(\nu_{\mathbf{k}} + l) = F_{\mathbf{k},n+l}^{10}(\nu_{\mathbf{k}}) \quad (\text{A5})$$

$$G_{\mathbf{k},n}(\nu_{\mathbf{k}} + l) = G_{\mathbf{k},n+l}(\nu_{\mathbf{k}}). \quad (\text{A6})$$

This naturally signifies that the system's behavior is entirely defined by its values within the Floquet-Brillouin zone.

A solution exists for the system of homogeneous equations (A1)-(A3) if the determinant of $\mathfrak{M} + \omega\nu_{\mathbf{k}}\mathbb{1}$ zero (cf. equation 12). In instances where the Floquet exponent is not an integer, it is possible to normalize the rows of this infinite-dimensional matrix in such a way that the diagonal elements become unity. After employing the particle-hole symmetry, we subsequently obtain a matrix that follows the pattern

$$\mathfrak{M} = \mathbb{1} + \begin{pmatrix} \dots & \dots & \dots & \dots & \dots \\ \dots & D_{n-1} & 0 & 0 & \dots \\ \dots & 0 & D_n & 0 & \dots \\ \dots & 0 & 0 & D_{n+1} & \dots \\ \dots & \dots & \dots & \dots & \dots \end{pmatrix} \\ + \begin{pmatrix} \dots & \dots & \dots & \dots & \dots \\ \dots & C_{n-1,0} & C_{n-1,+1} & C_{n-1,+2} & \dots \\ \dots & C_{n,-1} & C_{n,0} & C_{n,+1} & \dots \\ \dots & C_{n+1,-2} & C_{n+1,-1} & C_{n+1,0} & \dots \\ \dots & \dots & \dots & \dots & \dots \end{pmatrix} \quad (\text{A7})$$

with the 3×3 matrices

$$C_{n,m} = \begin{pmatrix} 0 & -\frac{T_{\mathbf{k},m}^{\parallel}}{\omega(n+\nu_{\mathbf{k}})+U} & 0 \\ -\frac{T_{\mathbf{k},m}^{\parallel}}{2\omega(n+\nu_{\mathbf{k}})} & 0 & \frac{T_{\mathbf{k},m}^{\parallel}}{2\omega(n+\nu_{\mathbf{k}})} \\ 0 & \frac{T_{\mathbf{k},m}^{\parallel}}{\omega(n+\nu_{\mathbf{k}})-U} & 0 \end{pmatrix} \quad (\text{A8})$$

and

$$D_n = \begin{pmatrix} 0 & -\frac{T_{\mathbf{k}}^{\perp}}{\omega(n+\nu_{\mathbf{k}})+U} & 0 \\ -\frac{T_{\mathbf{k}}^{\perp}}{2\omega(n+\nu_{\mathbf{k}})} & 0 & \frac{T_{\mathbf{k}}^{\perp}}{2\omega(n+\nu_{\mathbf{k}})} \\ 0 & \frac{T_{\mathbf{k}}^{\perp}}{\omega(n+\nu_{\mathbf{k}})-U} & 0 \end{pmatrix} \quad (\text{A9})$$

The determinant can be computed perturbatively in the limit of strong interactions and we find

$$0 \stackrel{!}{=} \text{Det}(\mathfrak{M}) = 1 - \frac{1}{2} \left[\sum_n \text{Tr}(D_n^2) + 2 \sum_n \text{Tr}(D_n C_{n,0}) \right. \\ \left. + \sum_{n,m} \text{Tr}(C_{n,m} C_{n+m,-m}) \right] + \mathcal{O}(T^4). \quad (\text{A10})$$

Upon the evaluation of traces and summations, we ultimately reach equation (21).

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