Missing quantum number of Floquet states

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We reformulate the Floquet theory for periodically driven quantum systems following a perfect analogy with the proof of the Bloch theorem. We observe that the current standard method for calculating the Floquet eigenstates using the quasienergy alone is incomplete and unstable and pinpoint an overlooked quantum number, the average energy. This new quantum number resolves many shortcomings of the Floquet method stemming from the quasienergy degeneracy issues, particularly in the continuum limit. Using the average-energy quantum number, we get properties similar to those of the static energy, including a unique lower-bounded ordering of the Floquet states, from which we define a ground state, and a variational method for calculating the Floquet states. This is a first step towards reformulating Floquet first-principles methods, which have long been thought to be incompatible due to the limitations of the quasienergy.

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I. INTRODUCTION

Periodically driven quantum systems have been gaining increasing attention, particularly due to the promise of Floquet engineering [1,2] to control material properties and achieve exotic states such as laser-induced superconductivity [3–5] and topological insulators [6,7]. So far these applications have been investigated only on simple model systems due to the limitations of the current Floquet methodology. In order to extend the Floquet formalism to self-consistent first-principles calculations and bigger, more complex systems, we need to reevaluate the source of these limitations.

Since the original derivations of the Floquet formalism in quantum mechanics [8–10], the fundamental methodology of solving the time-periodic Schrödinger equation using Floquet theory has remained unchanged, relying on the definition of the quasienergy eigenstates defined by the eigenproblem

$$\hat{H}^F(t)|\Phi_n(t)\rangle = \epsilon_n|\Phi_n(t)\rangle, \tag{1}$$

where $\hat{H}^F$, Φ, and ϵ denote the Floquet Hamiltonian [$\hat{H}^F(t) = \hat{H}(t) - i\hat{\partial}_t$], the time-periodic Floquet states [$\Phi(t + T) = \Phi(t)$], and the quasienergies, respectively. However, we know that there are various limitations to this definition [11–13]: the quasienergy ordering being meaningless, the breakdown of this eigenproblem in the continuum system, the lack of a Hilbert-space truncation method, and so on. These limitations are currently the main reasons why the Floquet formalism has not been generalized to various first-principles calculation methods [14–16].

As for the similarities between the Floquet and Bloch theories, they have long been known [1,17], but a detailed analogy between the two theories has not been thoroughly explored. If we explore this analogy (Table I), we find that the current Floquet formalism is apparently incomplete, using only one quantum number to label the eigenstates as opposed to the two in Bloch systems. It should be clarified that the quasienergy ϵ is analogous to the crystal momentum $\vec{k}$, as opposed to the common misconception that it corresponds to the static energy E. Additionally, the Floquet method lacks a systematic derivation of the Floquet eigenstates from the fundamental symmetry of the time-periodic Hamiltonian and discrete time-translation operator, which is otherwise well established in the Bloch theory [18]. Here we will present this missing derivation, completing the Floquet picture and Floquet-Bloch analogy, showing that the average energy [19], our missing quantum number (QN), is the analog of the static energy, and as such, we can expect it to fulfill similar roles.

In Sec. II, we rederive the Floquet eigenstates from first principles, following the same steps as those in Bloch systems [18]. We discuss the main differences between this method and the conventional Floquet approach in Sec. III and then the properties of the average energy as a QN in Sec. IV. A simple example of the two-level system illustrating these concepts is given in Sec. V. We conclude in Sec. VI, where we discuss some potential applications following this reformulation.

II. REDERIVING THE FLOQUET EIGENSTATES

First, we have to bring the time-periodic problem to a Hilbert space similar to that in the spatially periodic system. For this we promote the time parameter t to an operator defined on a Lebesgue space over the whole real domain $\mathbb{E} = L^2(\mathbb{R}, t)$. The time-dependent Hamiltonian is then defined as a self-adjoint operator on the extended Hilbert space $\mathbb{H} \otimes \mathbb{E}$, where $\mathbb{H}$ is the Hilbert space upon which the Hamiltonian

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TABLE I. Perfect analogy of Floquet theory and Bloch's theorem.

		Floquet theory	
	Bloch theorem	Quasienergy only	With average energy
Eigenstates	$(u_{\vec{k}n}(\vec{r}), \vec{k}, E_{\vec{k}n})$	$(\Phi_n(t), \epsilon_n)$	$(\Phi_{na}(t), \epsilon_n, \bar{E}_{na})$
Symmetry QN	$\hat{\vec{k}}\|u_{\vec{k}n}\rangle = \vec{k}\|u_{\vec{k}n}\rangle$	$\hat{H}^F\|\Phi_n\rangle = \epsilon_n\|\Phi_n\rangle$	$\hat{H}^F\|\Phi_{na}\rangle = \epsilon_n\|\Phi_{na}\rangle$
Ordering QN	$\hat{H}_{\vec{k}}\|u_{\vec{k}n}\rangle = E_{\vec{k}n}\|u_{\vec{k}n}\rangle$		$\hat{\bar{H}}_n\|\Phi_{na}\rangle = \bar{E}_{na}\|\Phi_{na}\rangle$
Ritz variation	$E[u] \geqslant E_0$	$\nexists$	$\bar{E}[\Phi] \geqslant \bar{E}_0$

$\hat{H}(t)$ acts at any time t. In this representation, the state vectors are expressed as

$$|\Psi\rangle = \int_{-\infty}^{+\infty} |\Psi(t)\rangle \otimes |t\rangle dt, \tag{2}$$

$$\langle\Psi|\Psi'\rangle = \int_{-\infty}^{+\infty} \langle\Psi(t)|\Psi'(t)\rangle dt, \tag{3}$$

and the time-periodic Hamiltonian is naturally extended to the following form:

$$\hat{H} = \int_{-\infty}^{+\infty} \hat{H}(t) \otimes |t\rangle\langle t| dt. \tag{4}$$

The physicality of this Hilbert-space extension is a topic of great debate [20], with the most prominent counterpoint being that the extended Hilbert space $\mathbb{H} \otimes \mathbb{E}$ is overcomplete [21,22], including unphysical states which do not satisfy the time-dependent Schrödinger equation. We overcome this issue by focusing on the physical subspace $(\mathbb{H} \otimes \mathbb{E})_S$ [23,24], defined as

$$\Psi \in (\mathbb{H} \otimes \mathbb{E})_S \quad \Leftrightarrow \quad [\hat{H} - i\hat{\partial}_t]|\Psi\rangle = 0|\Psi\rangle, \tag{5}$$

where the time-derivative operator $\hat{\partial}_t$ is expressed in the $\mathbb{E}$ Hilbert space as

$$\hat{\partial}_t = \int_{-\infty}^{+\infty} |t\rangle \partial_t \langle t| dt. \tag{6}$$

Defining the physical subspace as such can be intimidating, so instead we use the equivalence of the time-dependent Schrödinger equation with the Floquet Schrödinger equation; that is, any eigensolution of Eq. (1) corresponds to a physical state in $(\mathbb{H} \otimes \mathbb{E})_S$:

$$|\Psi_n(t)\rangle = e^{-i\epsilon_n t}|\Phi_n(t)\rangle. \tag{7}$$

Here we use the already established identities of the quasienergy eigenstates [1,9] but refrain from defining them as the final Floquet eigenstates. For the sake of completeness, we include the original derivations of these eigenstates in Appendix A. As our goal is not to deny the validity of this definition, but rather to complete it, we will use the quasienergy eigenbasis to simplify some identities. Therefore, we can express the projection operator $\mathbb{1}_S$ onto the physical subspace $(\mathbb{H} \otimes \mathbb{E})_S$ using the quasienergy basis set as

$$\mathbb{1}_S = \sum_n |\Psi_n\rangle\langle\Psi_n|, \tag{8}$$

where we require the orthonormality condition

$$\langle\Psi_m|\Psi_n\rangle = \delta_{mn}, \tag{9}$$

or, more explicitly,

$$|\Psi_n\rangle = \lim_{\mathcal{T}\to\infty} \frac{1}{\sqrt{2\mathcal{T}}} \int_{-\mathcal{T}}^{\mathcal{T}} e^{-i\epsilon_n t}|\Phi_n(t)\rangle \otimes |t\rangle dt. \tag{10}$$

For simplicity we have taken the Hilbert space $\mathbb{H}$ to be finite, although the generalization to the continuous space is straightforward. In order for the projection $\mathbb{1}_S$ to be unitary and complete in $(\mathbb{H} \otimes \mathbb{E})_S$, the summation label n in Eq. (8) is limited to a single quasienergy Brillouin zone ($\epsilon_n \in [0, \omega)$).

Next, in the Hilbert space $\mathbb{E}$ we define the time-translation operator $\hat{T}$ that shifts the time parameter by a time period T:

$$\hat{T} = \int_{-\infty}^{+\infty} |t - T\rangle\langle t| dt. \tag{11}$$

This operator trivially commutes with the time-periodic Hamiltonian $\hat{H}$ in the extended space $\mathbb{H} \otimes \mathbb{E}$ [Eq. (4)],

$$[\hat{H}, \hat{T}] = 0, \tag{12}$$

and it is the equivalent starting point of the Bloch theorem proof [18]. However, for our purposes, we need a similar commutation relation to hold within the physical subspace $(\mathbb{H} \otimes \mathbb{E})_S$. For that we project and redefine these operators on the physical subspace and confirm that the commutation relation still holds there.

The projection of the Hamiltonian $\hat{H}$ on the physical subspace gives us the average-energy operator $\hat{\bar{H}}$:

$$\hat{\bar{H}} = \mathbb{1}_S \hat{H} \mathbb{1}_S = \sum_{mn} \bar{H}_{mn}|\Psi_m\rangle\langle\Psi_n|, \tag{13}$$

$$\bar{H}_{mn} = \lim_{\mathcal{T}\to\infty} \frac{1}{2\mathcal{T}} \int_{-\mathcal{T}}^{\mathcal{T}} \langle\Psi_m(t)|\hat{H}(t)|\Psi_n(t)\rangle dt. \tag{14}$$

We refer to this operator as the average energy because its expectation value gives us the observable average energy $\bar{E}$, as defined in [19], for any normalized physical wave function in $(\mathbb{H} \otimes \mathbb{E})_S$:

$$\bar{E}[\Psi] = \langle\Psi|\hat{\bar{H}}|\Psi\rangle = \lim_{\mathcal{T}\to\infty} \frac{1}{2\mathcal{T}} \int_{-\mathcal{T}}^{\mathcal{T}} \langle\Psi(t)|\hat{H}(t)|\Psi(t)\rangle dt. \tag{15}$$

Using the definition of Eq. (10) in Eq. (14), we can see that the average-energy operator $\hat{\bar{H}}$ is diagonal with respect to different quasienergies, and it generally lifts the quasienergy degeneracy, so that we can simplify it to the following form:

$$\bar{H}_{mn} = \begin{cases} 0 & \text{if } \epsilon_m \neq \epsilon_n, \\ \dfrac{1}{T}\displaystyle\int_0^T \langle\Phi_m(t)|\hat{H}(t)|\Phi_n(t)\rangle dt & \text{if } \epsilon_m = \epsilon_n. \end{cases} \tag{16}$$

As for the translation operator $\hat{T}$, we can quickly see that it is diagonal with respect to the quasienergy basis defined in Eqs. (8) and (10):

$$\hat{T}_S = \mathbb{1}_S \hat{T} \mathbb{1}_S = \sum_n e^{-i\epsilon_n T} |\Psi_n\rangle\langle\Psi_n|. \tag{17}$$

From the decompositions and identities in Eqs. (13), (16), and (17), we can conclude that the average-energy operator $\hat{\bar{H}}$ and the time-translation operator $\hat{T}_S$ commute nontrivially in the physical subspace $(\mathbb{H} \otimes \mathbb{E})_S$ (see Appendix B for the proof). We thus define the physical eigenstates Ψ_{na} as the simultaneous eigenstates of these operators, with n being the QN of the quasienergy ϵ_n and a being the additional QN of the average-energy eigenvalue $\bar{E}_{na}$:

$$[\hat{\bar{H}}, \hat{T}_S] = 0, \tag{18}$$

$$\hat{T}_S |\Psi_{na}\rangle = e^{-i\epsilon_n T} |\Psi_{na}\rangle, \tag{19}$$

$$\hat{\bar{H}} |\Psi_{na}\rangle = \bar{E}_{na} |\Psi_{na}\rangle. \tag{20}$$

This definition of the physical eigentriplet $(\Psi_{na}, e^{-i\epsilon_n T}, \bar{E}_{na})$ is the first major result we want to emphasize in this work.

Finally, we substitute the Floquet state identity [Eq. (7)] of the eigenstates

$$|\Psi_{na}(t)\rangle = e^{-i\epsilon_n t} |\Phi_{na}(t)\rangle, \tag{21}$$

into Eqs. (19) and (20) to get the equivalent eigenproblem in the Floquet space $\mathbb{H} \otimes \mathbb{T}$, where $\mathbb{T} = L^2([0, T], t)$ is the Fourier space,

$$e^{i\epsilon_n t} \hat{T}_S e^{-i\epsilon_n t} |\Phi_{na}(t)\rangle = e^{-i\epsilon_n T} |\Phi_{na}(t)\rangle, \tag{22}$$

$$e^{i\epsilon_n t} \hat{\bar{H}} e^{-i\epsilon_n t} |\Phi_{na}(t)\rangle = \bar{E}_{na} |\Phi_{na}(t)\rangle. \tag{23}$$

Equation (22) is replaced with the usual quasienergy Floquet eigenproblem [Eq. (1)], and Eq. (23) gives us the equivalent average-energy operator $\hat{\bar{H}}_n$ acting on the Floquet space $\mathbb{H} \otimes \mathbb{T}$ (see Appendix C for the derivation),

$$\hat{\bar{H}}_n = \sum_{ij} \bar{H}_{nij} |\Phi_{ni}\rangle\langle\Phi_{nj}|, \tag{24}$$

$$\bar{H}_{nij} = \frac{1}{T} \int_0^T \langle\Phi_{ni}(t)|\hat{H}(t)|\Phi_{nj}(t)\rangle dt, \tag{25}$$

where i and j are the degenerate labels spanning the quasienergy degenerate subspace:

$$\Phi_{ni} \in (\mathbb{H} \otimes \mathbb{T})_{\epsilon_n} \quad \Leftrightarrow \quad \hat{H}^F |\Phi_{ni}\rangle = \epsilon_n |\Phi_{ni}\rangle. \tag{26}$$

At this point we should recall the labeling convention used here in order to not create any confusion with the different labeling conventions used in Floquet systems. Here we use m and n for the quasienergy labels, i and j for the quasienergy degenerate labels, and a and b for the average-energy labels, which are a subset of the former. We will also point out that the quasienergy ϵ_n dependence of the average-energy operator $\hat{\bar{H}}_n$ is identical to the crystal momentum $\vec{k}$ dependence of the effective Hamiltonian $\hat{H}_{\vec{k}}$ of the Bloch systems.

Putting it all together, we have the fundamental commutation relation

$$[\hat{H}^F, \hat{\bar{H}}_n] = 0, \tag{27}$$

from which we redefine the Floquet eigenstates Φ_{na} to be the simultaneous eigenstates of both the Floquet Hamiltonian $\hat{H}^F$ and the average-energy operator $\hat{\bar{H}}_n$, having the quantum numbers of the quasienergy ϵ_n and average energy $\bar{E}_{na}$, respectively. Notice the addition of Eq. (29) to this Floquet eigenstate definition:

$$\hat{H}^F |\Phi_{na}\rangle = \epsilon_n |\Phi_{na}\rangle, \tag{28}$$

$$\hat{\bar{H}}_n |\Phi_{na}\rangle = \bar{E}_{na} |\Phi_{na}\rangle. \tag{29}$$

III. DIFFERENCE FROM THE CONVENTIONAL FLOQUET METHOD

Conceptually, the conventional Floquet method of calculating the quasienergy eigenstates is analogous to calculating the Brillouin zone in spatially periodic Bloch systems, and the additional step proposed here in Eq. (29) is analogous to then calculating the energy bands' structure. We can imagine the significance of this step using this analogy, but to be more concrete, we will explicitly explore in this section the two main consequences of this redefinition. In the next section we will further justify it by looking at the properties of the average energy.

First, it resolves the quasienergy degeneracy. Traditional Floquet eigenstates are ill-defined within the quasienergy degenerate subspace, so that any rotated basis set $\{\Phi'_{nj}\}$, where

$$|\Phi'_{nj}\rangle = \sum_i C_{ij} |\Phi_{ni}\rangle, \tag{30}$$

$$\langle\Phi'_{ni}|\Phi'_{nj}\rangle = \delta_{ij}, \tag{31}$$

is an equally valid eigenbasis

$$\hat{H}^F |\Phi'_{nj}\rangle = \epsilon_n |\Phi'_{nj}\rangle \quad \forall \{\Phi'_{nj}\}. \tag{32}$$

This makes it impossible to uniquely define the quasienergy eigenstates in the continuum limit, and numerical calculations for these states become unstable. Conventionally, one would use an adiabatic continuation or perturbative method to resolve this ambiguity; however, the former is ill defined in the continuum limit [11], and the latter is unstable against infinitesimal perturbation [25]. The average energy offers an alternative labeling method that is much more efficient than the previously mentioned ones because it requires a single calculation step, and it is applicable in the continuum system. We comment that the current semiadiabatic Floquet methods [11,26] are equivalent and more intuitive if we perform the adiabatic continuation using the average energy [25].

Second, this labeling is robust against infinitesimal perturbations. It is natural to assume that a physical system would be unaffected by infinitesimally small perturbations within acceptable measurement constraints; that is, if we take $\hat{H}^0(t)$ to be an unperturbed Hamiltonian and $v(t)$ to be an arbitrary infinitesimally small perturbation, we expect that

$$|\Phi_n\rangle \approx |\Phi_n^0\rangle, \tag{33}$$

where the standard notations for the perturbed and unperturbed systems are implied. This assumption, however, fails around the degeneracy, where

$$\exists\, \hat{v}(t): \quad \big||\Phi_{ni}\rangle - |\Phi_{nj}^0\rangle\big| = O(1) \quad \forall\, i, j. \tag{34}$$

It is thus evident how problematic it is when we consider the continuum system, where we have infinitely dense degeneracies, and how numerical computations become unstable.

On the other hand, if we expand the derivation given here to include the near degeneracies, the labeling of the eigenstates becomes stable again [25], i.e.,

$$|\Phi_{na}\rangle \approx \left|\Phi^0_{na}\right\rangle, \quad \epsilon_n \approx \epsilon^0_n, \quad \bar{E}_{na} \approx \bar{E}^0_{na}. \tag{35}$$

The average energy $\bar{E}$ here is redefined to be the time average over a finite time. More discussions on this topic can be found in our previous works [19,25].

Outside the quasienergy degeneracies, there are virtually no differences in the Floquet eigenstates from the conventional Floquet method. Yet there are unique properties that the average energy brings as a quantum number that we should consider.

IV. PROPERTIES OF THE AVERAGE ENERGY

First, all of the familiar theorems related to the energy eigenstates, such as the Hellmann-Feynman theorem, have analogs with respect to both the quasienergy and the average energy, as they follow straight from the eigendefinition in Eqs. (28) and (29). Although the quasienergy ones were already known [9], because of the infinitely dense quasienergy degeneracy in the continuum, the usefulness of these theorems is greatly diminished. On the other hand, upon including the average energy, these degeneracies are lifted, and we can more naturally apply these methodologies to the continuum system.

Here we want to highlight that now a Ritz variational principle is possible:

$$\bar{E}[\Phi] = \langle\Phi|\sum_n \hat{\bar{H}}_n|\Phi\rangle \geqslant \bar{E}_0 \quad \forall\, \Phi \in \mathbb{H}\otimes\mathbb{T}, \tag{36}$$

where the equality sign occurs at the Floquet ground state. The lower boundness of $\bar{E}_0$ is guaranteed if the Hamiltonian is lower bounded at all times t. However, Eq. (36) is impractical in this form, as it would require the prior calculation of the full quasienergy eigenspectra through the definition of Eq. (24), and it is given here as such only for theoretical purposes. In practice we can exchange it with a more calculable effective average-energy functional $\bar{\mathcal{E}}[\Phi]$ [25] (see Appendix D for proof), defined as

$$\bar{\mathcal{E}}[\Phi] = \frac{1}{T}\int_0^T \langle\Phi(t)|\hat{H}(t)|\Phi(t)\rangle dt, \tag{37}$$

which becomes equivalent to the average energy $\bar{E}[\Phi]$ when the quasienergy $\epsilon[\Phi]$ is stationary,

$$\bar{\mathcal{E}}[\Phi] = \bar{E}[\Phi] \quad \Leftrightarrow \quad \Phi \in \{\Phi' \in \mathbb{H}\otimes\mathbb{T} \,|\, \delta\epsilon[\Phi'] = 0\}, \tag{38}$$

$$\epsilon[\Phi] = \frac{1}{T}\int_0^T \langle\Phi(t)|[\hat{H}(t) - i\partial_t]|\Phi(t)\rangle dt. \tag{39}$$

So, in practice, the efficient Lagrangian minimization method becomes

$$\bar{E}_0 = \min_{\Phi\in\mathbb{H}\otimes\mathbb{T}} \left\{\bar{\mathcal{E}}[\Phi] + \sum_i \lambda_i \frac{\delta\epsilon[\Phi]}{\delta\Phi_i} + \mu(\langle\Phi|\Phi\rangle - 1)\right\}, \tag{40}$$

where $\{\Phi_i\}$ is an arbitrary basis spanning the Floquet space $\mathbb{H}\otimes\mathbb{T}$. We can thus straightforwardly calculate the ground state by constraint minimization methods in the Floquet space $\mathbb{H}\otimes\mathbb{T}$ using only easily calculable functionals. This offers an alternative to the conventional perturbation and adiabatic continuation method, and it does not require any approximation to the Floquet Hamiltonian $\hat{H}^F$, just adequate parametrization of the Floquet wave function.

Second, the average energy is robust against infinitesimal perturbations. However, this was already mentioned in Sec. III. Here we will add that, with the lower-bounded and unconfined ordering of the eigenstates Φ_{na} by the average energy $\bar{E}_{na}$, we can now systematically truncate the Floquet space $\mathbb{H}\otimes\mathbb{T}$ to a computationally accessible subspace, focusing on the lowest average-energy states. This method would not require any adiabatic continuation to figure out which Floquet states are significant or the prior evaluations of the static energy eigenstates and is thus more computationally efficient and more generally applicable.

Third, the thermalization process of closed Floquet systems has to be revisited in light of the Floquet eigenstate redefinition using the average energy as the main quantum number. Currently, it is understood, and often overly generalized, that any state of a nonintegrable Floquet system heats up to a featureless infinite-temperature state [27] due to the Brillouin zone being infinitely dense with degeneracies and the quasienergy eigenstates having a vanishing convergence radius. However, these issues are resolved by the addition of the average energy as a quantum number and by its approximate form, respectively. Particularly, the minimization process in Eq. (40) gives us the stable states with finite average energies which do not "heat up" to infinite temperature, as its propagation is well defined by Eq. (7) absent of an arbitrary mixing of quasienergy eigenstates. This is in contrast to the arbitrarily chosen degenerate quasienergy basis or arbitrary superposition state which uncontrollably mixes the average-energy states.

We expect that more careful analysis of the thermalization process using the average energy would give us a more intuitive understanding of its mechanism, including in the special cases of the prethermal states [28,29]. But the thermalization of the average-energy Floquet eigenstates is a complex topic far beyond the scope of this paper, to which we will return to in future research.

Finally, as for the physical significance of the average energy, we expect that it can serve a role similar to the static energies in the thermal equilibrium. There have already been attempts to quantify the Floquet steady state by the average energy [30], although using a different definition for the average energy, showing how we cannot generally formulate an equivalent Boltzmann distribution around it. This is mostly due to the steady state being defined primarily by the energy spectra and its dependence on the detailed system-bath interaction. So instead we should consider the average energy as an approximation tool for systematically truncating the Hilbert space to a finite computable one. We can at the very least eliminate the highly excited states as long as the energy spectra are weakly overlapping [25].

V. EXAMPLE: TWO-LEVEL SYSTEM

The most straightforward example that demonstrates the difference of using this additional average-energy quantum number is in the driven two-level system with the time-dependent Hamiltonian

$$\hat{H}(t) = \begin{bmatrix} \frac{\omega_0}{2} & \frac{V}{2}e^{-i\omega t} \\ \frac{V}{2}e^{i\omega t} & -\frac{\omega_0}{2} \end{bmatrix}. \tag{41}$$

Here we use the standard notations for $\omega_0, V, \omega, \delta$, and Ω, representing the undriven energy difference, driving strength, driving frequency, detuning, and Rabi frequency, respectively. The last two are evaluated as

$$\delta = \omega - \omega_0, \quad \Omega = \sqrt{V^2 + \delta^2}. \tag{42}$$

We will assume that $\omega > \omega_0/2$ and focus on the resonance condition $\Omega = \omega$. Then we can easily construct an arbitrary degenerate eigenbasis $\{\Phi'_{\pm\alpha}\}$ with quasienergy $\epsilon_\pm = 0$:

$$\hat{H}^F(t)|\Phi'_{\pm\alpha}(t)\rangle = 0|\Phi'_{\pm\alpha}(t)\rangle, \tag{43}$$

$$|\Phi'_{+\alpha}(t)\rangle = \begin{pmatrix} \cos\alpha\sqrt{\frac{2\omega-\omega_0}{2\omega}} + \sin\alpha\sqrt{\frac{\omega_0}{2\omega}}e^{-i\omega t} \\ -\cos\alpha\sqrt{\frac{\omega_0}{2\omega}}e^{i\omega t} + \sin\alpha\sqrt{\frac{2\omega-\omega_0}{2\omega}} \end{pmatrix}, \tag{44}$$

$$|\Phi'_{-\alpha}(t)\rangle = \begin{pmatrix} \sin\alpha\sqrt{\frac{2\omega-\omega_0}{2\omega}} - \cos\alpha\sqrt{\frac{\omega_0}{2\omega}}e^{-i\omega t} \\ -\sin\alpha\sqrt{\frac{\omega_0}{2\omega}}e^{i\omega t} - \cos\alpha\sqrt{\frac{2\omega-\omega_0}{2\omega}} \end{pmatrix} \tag{45}$$

for any choice of $\alpha \in \mathbb{R}$. If we numerically calculate the Floquet eigenstates at this resonance condition, we will get an arbitrary basis of a value α and their Brillouin zone replicas. Focusing on a single Brillouin zone, we calculate the average-energy operator $\hat{\bar{H}}$ [Eq. (24)] as

$$\hat{\bar{H}} = \begin{bmatrix} -\frac{\omega_0}{2}\cos 2\alpha & -\frac{\omega_0}{2}\sin 2\alpha \\ -\frac{\omega_0}{2}\sin 2\alpha & \frac{\omega_0}{2}\cos 2\alpha \end{bmatrix}, \tag{46}$$

which all give the average-energy Floquet eigenstates

$$|\Phi_+(t)\rangle = \begin{pmatrix} \sqrt{\frac{2\omega-\omega_0}{2\omega}} \\ -\sqrt{\frac{\omega_0}{2\omega}}e^{i\omega t} \end{pmatrix}, \quad \bar{E}_+ = -\frac{\omega_0}{2}, \tag{47}$$

$$|\Phi_-(t)\rangle = \begin{pmatrix} \sqrt{\frac{\omega_0}{2\omega}}e^{-i\omega t} \\ \sqrt{\frac{\omega-\omega_0}{2\omega}} \end{pmatrix}, \quad \bar{E}_- = \frac{\omega_0}{2}. \tag{48}$$

In this simple system, it is trivial to confirm that the eigenvalues $\bar{E}_\pm$ equal the observable average energy calculated from the propagation of the initial wave functions $\Phi(0)$ according to Eq. (15). But a more promising sign is that these eigenstates are equivalent to the adiabatically connected Floquet eigenstates:

$$|\Phi_+(V,t)\rangle = \begin{pmatrix} \sqrt{\frac{\Omega+\delta}{2\Omega}} \\ -\sqrt{\frac{\Omega-\delta}{2\Omega}}e^{i\omega t} \end{pmatrix}, \tag{49}$$

$$|\Phi_-(V,t)\rangle = \begin{pmatrix} \sqrt{\frac{\Omega-\delta}{2\Omega}}e^{-i\omega t} \\ \sqrt{\frac{\Omega+\delta}{2\Omega}} \end{pmatrix}, \tag{50}$$

$$\epsilon_\pm(V) = \mp\frac{\Omega-\omega}{2}, \tag{51}$$

$$\bar{E}_\pm(V) = \mp\frac{1}{2}\left(\Omega - \frac{\delta\omega}{\Omega}\right). \tag{52}$$

This is naturally to be expected because the Hellmann-Feynman theorem is applicable to the average-energy Floquet eigenstates. The other main property we can show in this simple system is the applicability of the minimization principle in Eq. (40), which we will discuss in the remainder of this section.

First, we confirm that any quasienergy variational method, such as the minimization of the quasienergy variance [31]

$$\min_\Phi\{\langle\Phi|(\hat{H}^F)^2|\Phi\rangle - \epsilon[\Phi]^2\} = 0, \tag{53}$$

gives us an arbitrary Floquet state of Eqs. (44) and (45). And, indeed, we can see this in Fig. 1(b), where we track the convergence process of this minimization method towards a quasienergy eigenstate. To track how the trial Floquet wave function $\Phi(t)$ within the Floquet space $\mathbb{H}\otimes\mathbb{T}$ varies in this search, we follow the parameter θ derived from the corresponding wave function $\Phi(0)$ at $t = 0$ such that

$$\frac{|\Phi(0)\rangle}{\langle\Phi(0)|\Phi(0)\rangle} = \begin{pmatrix} \cos\theta \\ \sin\theta \end{pmatrix}. \tag{54}$$

The starting condition for each minimization search is taken to be the static wave function

$$|\Phi_0(t)\rangle = \begin{pmatrix} \cos\theta \\ \sin\theta \end{pmatrix}, \tag{55}$$

with θ being the value at iteration 0 in Fig. 1. The nonconverging searches are not plotted here, and we have confirmed that all of the final states correspond to either the respective wave functions in Eq. (44) or Eq. (45) or a shifted replica of them in a different Brillouin zone.

Next, the minimization processes in Eq. (40) with the same initial conditions are shown in Fig. 1(a). Here we see that the vast majority of calculations converge to the ground state Φ_+ or an equivalent replica in another Brillouin zone. A small minority of calculations get stuck at the local critical point of Φ_-, which is naturally expected in such a gradient approach. The agreement of the final converging wave functions with the analytic solutions in Eqs. (47) and (48) or their replicas is close to the 10^{-12} level of the average energy and constraint convergence criteria.

We thus see that such a minimization method reliably converges to the well-defined average-energy ground state and can be trivially extended to calculate subsequent excited states. These solutions are consistent with the adiabatically continued states, which is thanks to the fact that the average energy and the quasienergy are compatible quantum numbers.

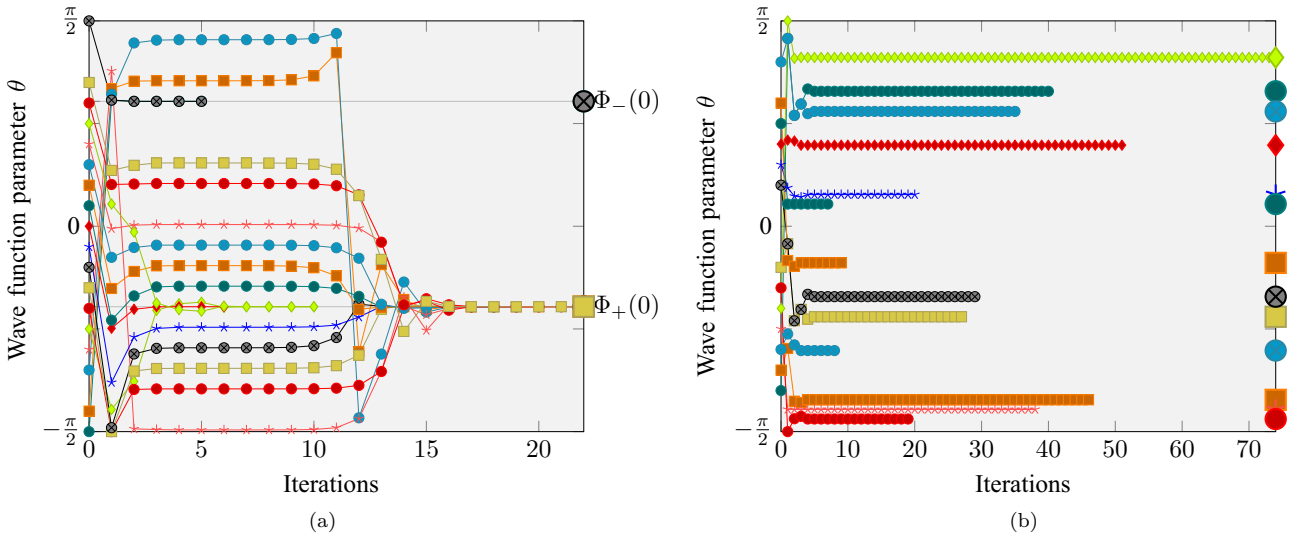

FIG. 1. Convergence to the Floquet states using the Lagrange minimization of (a) the average energy [Eq. (40)] and (b) the quasienergy variance [Eq. (53)]. Each line represents a different convergence path starting from the initial states described in Eq. (55), and the final converging solution is duplicated on the rightmost axes. Only the convergent solutions are plotted, where the convergence criteria for both the objective function and the constraints are set to a factor of 10^{-12}.

VI. CONCLUSION

To conclude, we reported that a more complete method for studying the time-periodic quantum systems using Floquet theory is to decompose the system into the Floquet eigenstates defined as the eigentriplet with the quasienergy and average energy. One of the main properties of this formulation is the introduction of the Ritz variational principle. Its lack has been a major roadblock in the development of efficient Floquet first-principles methods, which is most beautifully exemplified in the incompatibility of the Floquet density-functional theory [14,15]. Now, we can revisit these Floquet first-principles methods, which would bring us one step closer to efficiently simulating the steady state of a many-body electron system under constant laser irradiation. Indeed, the Floquet Hartree-Fock method is straightforward to derive [25].

The main physical significance of the average energy would be in calculating the steady state. We do not expect it to play exactly the same role as the static energy, and instead it should have a supporting one, such as limiting the Hilbert space to the physically significant states. To that end, the average-energy ground state might not have direct physical significance, but we conjecture that it generally approximates the steady state with dense thermal baths. Indeed we can check the validity of these statements in various model systems, while a quantitative proof is still being developed.

Finally, we should revisit the various ingrained understandings of the Floquet systems that have been based solely on the dynamics of the quasienergy eigenstates, such as the applicability of the eigenstate thermalization hypothesis. Here we provided a more robust definition of the Floquet eigenstates free from the infinite degeneracy issues of the previous definition, which gives a more intuitive understanding of various phenomena, such as the prethermalized states. We look forward to seeing research revisiting these well-established Floquet concepts and finding how they hold up without the infinite-degeneracy problem.

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APPENDIX A: ORIGINAL FORMULATION OF THE QUASIENERGY EIGENSTATES

The original derivation of the Floquet formalism [1,8] had only one goal of solving the time-dependent Schrödinger equation with the most general form,

$$i\partial_t \hat{U}(t,0) = \hat{H}(t)\hat{U}(t,0), \tag{A1}$$

where $\hat{U}$ is the usual propagator operator. Applying Floquet theory to Eq. (A1), we get the following identity of the propagator:

$$\hat{U}(t+T,0) = \hat{U}(t,0)\hat{U}(T,0), \tag{A2}$$

which we use to separate the micromotion $\hat{U}(t,0)$ from the Floquet Hamiltonian effects generalized by $\hat{U}(T,0)$. The latter is decomposed into its eigenstates, with the quasienergy exponents as eigenvalues:

$$\hat{U}(T,0) = \sum_n e^{-i\epsilon_n T}|\Phi_n(0)\rangle\langle\Phi_n(0)|. \tag{A3}$$

Combined with the micromotion, this gives us the well-established propagator form of the time-periodic propagator,

$$\hat{U}(t,0) = \sum_n e^{-i\epsilon_n t}|\Phi_n(t)\rangle\langle\Phi_n(0)|, \tag{A4}$$

and all of its related identities such as Eq. (1).

But here we note that the only guaranteed property of the quasienergy eigenstates $\Phi_n(t)$ and their superposition states is that they satisfy the time-dependent Schrödinger equation.

There is no mechanism introduced in this formulation to select or distinguish among these states the different dynamics of a stable, slowly propagating state from the myriad of highly excited states. This original derivation is not linked to any observable from which we can deduce the physical significance of these states beyond the relation in Eq. (A4) and its derivatives.

APPENDIX B: PROOF OF $[\hat{\bar{H}}, \hat{T}_S] = 0$

We can rewrite the operators $\hat{\bar{H}}$ and $\hat{T}_S$ as

$$\hat{\bar{H}} = \sum_{nij} \bar{H}_{nij} |\Psi'_{ni}\rangle\langle\Psi'_{nj}|, \tag{B1}$$

$$\hat{T}_S = \sum_{ni} e^{-i\epsilon_n T} |\Psi'_{ni}\rangle\langle\Psi'_{ni}| \tag{B2}$$

using an arbitrary quasienergy eigenbasis $\{\Psi'_{ni}\}$ [Eq. (26)], for which we will reserve the primed notation here for clarity. This basis satisfies

$$|\Psi'_{ni}\rangle = \lim_{\mathcal{T}\to\infty} \frac{1}{\sqrt{2\mathcal{T}}} \int_{-\mathcal{T}}^{\mathcal{T}} e^{-i\epsilon_n t} |\Phi'_{ni}(t)\rangle \otimes |t\rangle dt, \tag{B3}$$

$$[\hat{H}(t) - i\partial_t] |\Phi'_{ni}(t)\rangle = \epsilon_n |\Phi'_{ni}(t)\rangle, \tag{B4}$$

so that we retain the orthonormality conditions

$$\langle\Psi'_{mi}|\Psi'_{nj}\rangle = \delta_{mn}\delta_{ij}. \tag{B5}$$

Combining this orthonormality condition with the definitions in Eqs. (B1) and (B2), we straightforwardly get

$$\left[\hat{\bar{H}}, \hat{T}_S\right] = \sum_{mnijk} [\bar{H}_{mij} e^{-i\epsilon_n T} \delta_{mn}\delta_{jk} - e^{-i\epsilon_m T} \bar{H}_{njk}\delta_{mn}\delta_{ij}] |\Psi'_{mi}\rangle\langle\Psi'_{nk}| \tag{B6}$$

$$= \sum_{nij} [\bar{H}_{nik} e^{-i\epsilon_n T} - e^{-i\epsilon_n T} \bar{H}_{nik}] |\Psi'_{ni}\rangle\langle\Psi'_{nk}| = 0. \tag{B7}$$

The commutation of the effective operator $\hat{\bar{H}}_n$ and the Floquet Hamiltonian $\hat{H}^F$ in the Floquet space follows the same steps.

APPENDIX C: DERIVATION OF $\bar{H}_n$

Starting from the physical eigenstate definition,

$$\hat{\bar{H}} |\Psi_{na}\rangle = \bar{E}_{na} |\Psi_{na}\rangle, \tag{C1}$$

$$|\Psi_{na}\rangle = \lim_{\mathcal{T}\to\infty} \frac{1}{\sqrt{2\mathcal{T}}} \int_{-\mathcal{T}}^{\mathcal{T}} e^{-i\epsilon_n t} |\Phi_{na}(t)\rangle \otimes |t\rangle dt, \tag{C2}$$

we operate $\langle t|$ on the left of Eq. (C1), where t is an arbitrary time:

$$\langle t|\hat{\bar{H}}|\Psi_{na}\rangle = \bar{E}_{na}\langle t|\Psi_{na}\rangle. \tag{C3}$$

We expand the definition of $\hat{\bar{H}}$ from Eq. (B1),

$$\sum_{mij} \bar{H}_{mij}\langle t|\Psi'_{mi}\rangle\langle\Psi'_{mj}|\Psi_{na}\rangle = \bar{E}_{na}\langle t|\Psi_{na}\rangle. \tag{C4}$$

The inner product of the state vectors is simplified as

$$\langle\Psi'_{mj}|\Psi_{na}\rangle = \lim_{\mathcal{T}\to\infty} \frac{1}{2\mathcal{T}} \int_{-\mathcal{T}}^{\mathcal{T}} e^{i(\epsilon_m - \epsilon_n)t} \langle\Phi'_{mi}(t)|\Phi_{na}(t)\rangle dt \tag{C5}$$

$$= \delta_{mn} \frac{1}{T} \int_0^T \langle\Phi'_{mi}(t)|\Phi_{na}(t)\rangle dt. \tag{C6}$$

Now it is helpful to introduce a double bra-ket notation for the vectors in the Floquet space $\mathbb{H}\otimes\mathbb{T}$, such that the inner product is defined by

$$\langle\langle\Phi|\Phi'\rangle\rangle = \frac{1}{T}\int_0^T \langle\Phi(t)|\Phi'(t)\rangle dt \quad \forall\, \Phi, \Phi' \in \mathbb{H}\otimes\mathbb{T}. \tag{C7}$$

Using Eqs. (C6) and (C7) in Eq. (C4) and rearranging some terms, we get

$$\sum_{ij} \bar{H}_{nij} |\Phi'_{ni}(t)\rangle\langle\langle\Phi'_{nj}|\Phi_{na}\rangle\rangle = \bar{E}_{na}|\Phi_{na}(t)\rangle. \tag{C8}$$

As this relation holds at all times t, we can expand it to the whole Floquet space $\mathbb{T}$:

$$\left[\sum_{ij} \bar{H}_{nij} |\Phi'_{ni}\rangle\rangle\langle\langle\Phi'_{nj}|\right] |\Phi_{na}\rangle\rangle = \bar{E}_{na}|\Phi_{na}\rangle\rangle. \tag{C9}$$

Defining the bracketed expression as $\hat{\bar{H}}_n$, we get the original equation in Eq. (29). Repeating this process for all physical eigenstates Ψ_{na} and for all quasienergy Brillouin zones, we get the complete definition of $\hat{\bar{H}}_n$ spanning all Floquet space $\mathbb{H}\otimes\mathbb{T}$.

APPENDIX D: EQUIVALENCE OF $\bar{\mathcal{E}}$ AND $\bar{E}$

Expanding the functionals $\bar{E}[\Phi]$ and $\bar{\mathcal{E}}[\Phi]$ on an arbitrary quasienergy basis $\{\Phi'_{ni}\}$, we find the differences between them to be in the nondegenerate components with respect to the quasienergy labels, i.e.,

$$\bar{E}[\Phi] = \sum_{nij} C^*_{ni} C_{nj} \bar{H}_{nij}, \tag{D1}$$

$$\bar{\mathcal{E}}[\Phi] = \sum_{mnij} C^*_{mi} C_{nj} \bar{H}_{mnij}, \tag{D2}$$

where $\bar{H}_{mnij}$ is the more general uncontracted form of $\bar{H}_{nij}$ [Eq. (25)], representing the matrix element of the Hamiltonian,

$$\bar{H}_{mnij} = \frac{1}{T}\int_0^T \langle\Phi'_{mi}(t)|\hat{H}(t)\big|\Phi'_{nj}(t)\big\rangle dt, \tag{D3}$$

and C_{nj} is the usual overlap matrix with the arbitrary Floquet state Φ:

$$C_{nj} = \frac{1}{T}\int_0^T \langle \Phi'_{nj}(t)|\Phi(t)\rangle dt. \tag{D4}$$

So the condition for the two functionals to be equal simplifies to

$$\bar{E}[\Phi] = \bar{\mathcal{E}}[\Phi] \quad \Leftrightarrow \quad C^*_{mi}C_{nj} = \delta_{mn}C^*_{ni}C_{nj}. \tag{D5}$$

This condition is satisfied by the quasienergy variation $\delta\epsilon[\Phi] = 0$, which we will show explicitly here. For this we decompose the Floquet state Φ using two parameters, θ and φ, so that we focus on the two states Φ'_{mi} and Φ'_{nj} as follows:

$$|\Phi\rangle = \cos(\theta)\cos(\varphi)|\Phi'_{mi}\rangle + \cos(\theta)\sin(\varphi)|\Phi'_{nj}\rangle + \sin(\theta)|\Phi'^{\perp}\rangle. \tag{D6}$$

Here $\Phi'^{\perp}$ is the remaining projection of the Floquet state Φ orthogonal to both Φ'_{mi} and Φ'_{nj}. We will also ignore the complex phase as it does not affect the quasienergy variation. As the variation $\delta\epsilon[\Phi] = 0$ has to hold for all parameters, the following condition has to hold:

$$\frac{\delta\epsilon[\Phi]}{\delta\varphi} = -(\epsilon_m - \epsilon_n)\cos^2(\theta)\sin(2\varphi) = 0. \tag{D7}$$

So the necessary condition for the quasienergy variation to hold is

$$\delta\epsilon[\Phi] = 0 \;\Rightarrow\; \begin{cases} \epsilon_m = \epsilon_n & \Leftrightarrow\; m = n \quad \forall C_{mi}, C_{nj}, \\ \sin(2\varphi) = 0 & \Leftrightarrow\; C_{mi} = 0 \text{ or } C_{nj} = 0, \\ \cos(\theta) = 0 & \Leftrightarrow\; C_{mi} = C_{nj} = 0. \end{cases} \tag{D8}$$

Applying this for all pairs, we get the equivalence with Eq. (D5),

$$\delta\epsilon[\Phi] = 0 \quad \Leftrightarrow \quad C^*_{mi}C_{nj} = \delta_{mn}C^*_{ni}C_{nj}, \tag{D9}$$

and thus, Eq. (38) holds.

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