Semiconductor-Bloch Formalism: Derivation and Application to High-Harmonic Generation from Dirac Fermions

Jan Wilhelm, 1,* Patrick Grössing, 1 Jack Crewse, 1,2 Maximilian Nitsch, 1 Leonard Weigl, 3 Christoph Schmid, 3 and Ferdinand Evers 1,†

1 Institute of Theoretical Physics, University of Regensburg, Universitätsstraße 31, D-93053 Regensburg, Germany
2 Department of Physics, Missouri University of Science & Technology, Rolla, Missouri 65409, USA
3 Institute of Experimental and Applied Physics, University of Regensburg, Universitätsstraße 31, D-93053 Regensburg, Germany

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We rederive the semiconductor Bloch equations emphasizing the close link to the Berry connection. Our rigorous derivation reveals the existence of a third contribution to the (longitudinal) current in addition to the traditional intraband and polarization-related interband terms. The novel term becomes sizable in situations where the dipole-matrix elements are strongly wave-number dependent.

We apply the formalism to high-harmonic generation for a Dirac metal. The novel term adds to the frequency-dependent emission intensity (high-harmonic spectrum) significantly at certain frequencies reaching up to 90% of the total signal.

I. INTRODUCTION

The advancement of time-resolved spectroscopy seen in recent years facilitated the study of dynamical processes on sub-cycle time scales. Interesting effects that arise along the way in metals and semiconductors include the generation of high-harmonics (HHG) by the lightwave-driven charge carriers [1–7], subcycle control of charge transport in nanostructures [8], and atomic-resolution ultrafast microscopy [9]. Since high-harmonics are very sensitive to acceleration processes that the charge carriers are subjected to, HHG can be used for monitoring dynamical processes. Promising applications for band structure reconstruction [10, 11] and for observing dynamical Bloch oscillations [2,12] and Berry phase effects [13–15] have been reported.

An established theoretical framework to describe the dynamics of quantum systems is the density-matrix formalism that is known as semiconductor Bloch equations (SBE) in the context of crystalline solids. [16–21] It is exact, in principle, but in most applications of SBE, dynamical contributions from Coulomb interactions are neglected while band-structure effects are properly kept. This approximation has proven to be useful in numerous applications including, in particular, HHG in various model systems [15,22–27] and materials [28–32].

In the first part of the article, we present a rederivation of the main equations of motion (EoM) for the density matrix \( \rho(t) \) and the physical observables in the framework of SBE. The particular perspective we here offer emphasizes the close relation between SBE and the Berry connection.

Second, we present a rigorous derivation of the relation between \( \rho \) and the longitudinal current \( j(t) \). Our exact result reveals the existence of an additional term not accounted for in earlier work [33,34] and also not in most recent work by Baykusheva et al. [35] The consequences of this new term will be discussed. For the example of Dirac fermions, we find qualitative agreement with respect to the high-harmonic spectrum between the exact and the approximated expression. Quantitative discrepancies appear, however, which can exceed an order of magnitude. Implications of neglecting the extra term for most recent results [35] on high-harmonic generation in Dirac systems remain to be investigated.

The manuscript is organized as follows: Sec. II focuses on the EoM for the density matrix, \( \rho(t) \), with emphasis on SBE and the Berry connection. In Sec. III, we relate \( \rho(t) \) to the time-dependent longitudinal current density and the frequency-dependent emission intensity that underlies the HHG. An application to HHG in metallic films with Dirac-like spectrum is presented in Sec. IV.

II. DERIVATION OF EQUATIONS OF MOTION FOR THE DENSITY MATRIX

The power of the density matrix formalism is in its simplicity. In principle, it allows for the propagation of observables in a genuine many-body theory keeping the effective Hilbert space on the single-particle size. It thus can be intrinsically more efficient than wavefunction correlation theory [36–39]. Conceptually similar are Green’s function based approaches, such as GW + Bethe-Salpeter [40–45]. They keep an additional dynamical degree of freedom, however, and therefore tend to be computationally more expensive. An affordable alternative to density-matrix based approaches is the time-dependent density functional theory [46–62]. It has the advantage that implementations are available that can treat inhomogeneous systems of considerable size; progress towards including spatially varying electric fields
A. Equation of motion

Consider the fermionic, second quantized many-body Hamiltonian

\[ \hat{H} = \sum_{\alpha \beta} h_{\alpha \beta}(0)c_{\alpha}^{\dagger}c_{\beta} + \frac{1}{2} \sum_{\alpha, \beta, \gamma, \delta} U_{\alpha \beta \gamma \delta} c_{\alpha}^{\dagger}c_{\beta}^{\dagger}c_{\gamma}c_{\delta} \]  

(1)

with \( h(0) \) denoting a generic single-particle Hamiltonian represented in a stationary basis of a number of \( N_B \) single-particle states \( |\phi_\alpha^{(0)}\rangle \). In the presence of time-dependent perturbations, such as external electric or magnetic fields, this component of \( \hat{H} \) becomes time dependent \( h(t) \). The density matrix, \( \rho \), is defined by the matrix elements

\[ \rho_{\alpha \beta}(t) := \langle \Psi(t)|c_{\beta}^{\dagger}c_{\alpha}|\Psi(t)\rangle. \]  

(2)

It describes selected aspects of a time evolving many-body state \( \Psi(t) \) that enter physical observables, e.g., the particle density. The time-evolution of \( \rho(t) \) derives directly from the definitions Eqs. (1), (2) and the Schrödinger equation; in a basis-free representation the resulting EoM takes the form

\[ i\frac{\partial}{\partial t} \rho(t) = \{h(t), \rho\} + i \frac{\partial}{\partial t} \rho_{\text{coll}}(t), \]  

(3)

while the commutator in (3) accounts for the (effective) single-particle dynamics, the collision term \( \frac{\partial}{\partial t} \rho_{\text{coll}}(t) \) includes genuine two-body effects. Systematic expansions have been proposed to deal with it approximately, however, at the expense of a considerable numerical effort. [75, 76]

We here consider time-dependent Hartree-Fock theory, where the collision term is dropped and the time evolution of \( \rho \) remains unitary. In order to mimic the (non-unitary) effects of collisions, the collision term can be approximated on a heuristic level by replacing it with phenomenological damping terms [20]. For recent discussions on the strength and physical content of dephasing terms, we refer to Ref. 63.

The mean-field interaction, \( v_{\text{HF}} \), can be understood as a known functional of the density matrix \( v_{\text{HF}}(t) := \mathcal{F}[\rho] \),

\[ \mathcal{F}_{\alpha \beta}(t) := \sum_{\alpha \beta \gamma \delta} (U_{\alpha \gamma \beta \delta} - U_{\alpha \beta \gamma \delta}) \rho_{\gamma \delta}(t), \]  

(5)

where the matrix elements \( \rho_{\alpha \beta}(t) \) are the representation of \( \rho(t) \) in the stationary basis \( |\phi_\alpha(t)\rangle \). The functional (5) together with (3) gives a closed set of equations for the dynamics of \( \rho(t) \). Exchange-correlation functionals alternative to Eq. (5) have been explored in the spirit of (time-dependent) density functional theory. [77]

B. The adiabatic basis

We define an adiabatic basis [78] \( |\alpha; t\rangle \) by the simultaneous, orthonormalized eigenstates of \( h(t) \)

\[ h(t)|\alpha; t\rangle = \epsilon_\alpha(t)|\alpha; t\rangle. \]  

(6)

In this basis, the commutator dynamics (3) takes a simple form. Notice, that (6) defines the basis at time \( t \) only up to a phase factor. Therefore, two basis sets at neighboring times \( t \) and \( t+dt \), \( |\alpha; t\rangle \) and \( |\alpha; t+dt\rangle \), can differ, in principle, by an arbitrary phase factor so that the motion of matrix elements given in the adiabatic frame is not yet uniquely defined. We conclude that the time evolution of the phase-factor needs to be imposed by an extra condition that complements (6) but is not part of (6).

In order to formulate this condition we adopt the attitude that \( |\alpha; t\rangle \) and \( |\alpha; t+dt\rangle \) should be smoothly connected in a manner as it would be implied by perturbation theory; we thus stipulate

\[ \partial_t |\alpha; t\rangle := \sum_{\beta \neq \alpha} \langle \beta; t| \hat{h}(t)|\alpha; t\rangle \frac{\epsilon_\alpha(t) - \epsilon_\beta(t)}{\epsilon_\alpha(t) - \epsilon_\beta(t)} , \]  

(7)

which implies \( \langle \alpha; t|\partial_t|\alpha; t\rangle = 0 \). The time evolution (7) starts at \( t \to -\infty \) with initial eigenstates, \( |\alpha(-\infty)\rangle := |\phi_\alpha\rangle \), that are defined as

\[ \hat{h}\text{in}|\phi_\alpha\rangle = \epsilon_\alpha|\phi_\alpha\rangle, \quad \text{h}\text{in} := \lim_{t \to -\infty} h(t). \]  

(8)

To further connect the time evolution (7) to other definitions in the literature [78], we specify to a situation where \( h(t) \) is implicitly time dependent, because it contains a set of parameters \( \mathbf{R}(t) \) that are time dependent, \( h(\mathbf{R}(t)) \). These parameters could be, e.g., external electric or magnetic fields, \( \mathbf{E}(t) \) and \( \mathbf{B}(t) \), but in the case of self-consistent field theories also the matrix elements of \( \rho \) themselves. We thus have

\[ \partial_t |\alpha; t\rangle = \hat{R} \sum_{\beta \neq \alpha} \langle \beta; t| \frac{\partial}{\partial \mathbf{R}}|\alpha; t\rangle \frac{\epsilon_\alpha(t) - \epsilon_\beta(t)}{\epsilon_\alpha(t) - \epsilon_\beta(t)}. \]  

(9)
Suppressing the time-dependencies in our notation, the matrix element can be evaluated by observing that
\[
\partial_R \langle \beta | h | \alpha \rangle = \langle \partial_R \beta | h | \alpha \rangle + \langle \beta | \partial_R h | \alpha \rangle + \langle \beta | h | \partial_R | \alpha \rangle = \tilde{\epsilon}_\alpha \langle \partial_R \beta | \alpha \rangle + \langle \beta | \partial_R h | \alpha \rangle.
\]
Since \( \langle \beta | \partial_R | \alpha \rangle = -\langle \partial_R | \beta | \alpha \rangle \), we have
\[
\langle \beta | \partial_R h | \alpha \rangle = (\tilde{\epsilon}_\alpha - \tilde{\epsilon}_\beta) \langle \beta | \partial_R | \alpha \rangle + \delta_{\alpha \beta} \langle \beta | \partial_R | \alpha \rangle.
\]
When inserting this relation into (9) we arrive at the result
\[
\partial_t | \alpha; t \rangle := \hat{R}(t) \sum_{\beta \neq \alpha} | \beta; t \rangle \langle \beta; t | \partial_R | \alpha; t \rangle.
\]
We adopt the formulation of dynamics in the adiabatic basis as in Eq. (11) as our preferred one. It reveals the close connection to differential geometry, because it implies
\[
\hat{R}(t) | \alpha; t \rangle | \partial_R | \alpha; t \rangle = 0
\]
that we have obtained from \( \langle \alpha; t | \partial_t | \alpha; t \rangle = 0 \), see note below (7). Relation (12) is well known as the condition of parallel transport [78]; it is a result of the specific way to define the phase evolution of wave functions \( | \alpha; t \rangle \) during time by imposing (7). Eq. (12) implies that the motion of the adiabatic frame is such that the Berry connection [79]
\[
\mathcal{A}_\alpha[R] := \langle \alpha; t | i \partial_R | \alpha; t \rangle
\]
remains perpendicular to the "velocity" of each state \( | \alpha; t \rangle \).
\[
\hat{R}(t) \cdot \mathcal{A}_\alpha[R] = 0.
\]
We further illustrate the meaning of (11) discussing the example of Bloch electrons in homogeneous electric field.

**Bloch electrons in homogeneous \( E(t) \).** We consider charged free fermions, so \( \nu_{\text{HF}} \rightarrow 0 \) and \( h \rightarrow h^{(0)} \). They are embedded in a crystal lattice, so the eigenstates of the stationary single particle Hamiltonian (without electric field, \( E(t) = 0 \)) are Bloch-states \( | n k \rangle \), which implies \( | \alpha \rangle \rightarrow | n k \rangle \). Recalling Bloch’s theorem, we have a factorization of the eigenstates
\[
|r| n k \rangle = \frac{1}{\sqrt{N}} e^{i k r} (r| n k \rangle)
\]
with eigenvalues \( \epsilon_n(k) \); here, \( N \) denotes the number of unit cells and the matrix element on the rhs represent the lattice-periodic content of the Bloch state, \( u_{n k}(r) := (r| n k \rangle \) in a traditional notation [80]. The double angular brackets indicate that the normalization volume for \( u_{n k} \) is the unit cell, see Appendix A for more details on our notation. Formally, the states \( | n k \rangle \) are solutions of the eigenvalue problem
\[
h^{\text{in}}(k)| n k \rangle = \epsilon_n(k) | n k \rangle
\]
with
\[
h^{\text{in}}(k) := \sum_n | n k \rangle \epsilon_n(k) \langle n k |
\]
see Eq. (A13) in Appendix A. Summarizing, the stationary Bloch-Hamiltonian reads
\[
\hat{H} = \sum_{mm'} \int_{BZ} \frac{d k}{(2 \pi)^d} h^{\text{in}}_{mm'}(k) c_{m'}^\dagger(k) c_m(k)
\]
where \( h^{\text{in}}_{mm'}(k) := \langle m| h^{\text{in}}(k)| m' \rangle \) and the states \( | m \rangle \) denote a generic basis in the subspace of the degrees of freedom of the unit cell (bands) that may or may not be chosen to depend on \( k \).

As a time-dependent perturbation acting on fermions of charge \( q \), we introduce a homogeneous electric field \( E(t) \) that evolves from zero, i.e. \( \lim_{t \to -\infty} E(t) = 0 \). Its effect is discussed conveniently in the Coulomb gauge [81]
\[
q E(t) = -\dot{A}(t),
\]
where a factor \( q/c \) was absorbed in the definition of \( A \). As compared to the alternative gradient representation, \( E(t) = -\nabla \phi(r,t) \), the Coulomb gauge offers the advantage that it does not break translational invariance for homogeneous electric fields; therefore, it is particularly convenient for treating Bloch electrons. Using minimal coupling, we have [19, 78, 82]
\[
h(k; t) := h^{\text{in}}(k_t), \quad k_t = k - A(t)
\]
and correspondingly
\[
h^{\text{in}}(k_t)| n\sim k; t \rangle = \epsilon_n(k_t) | n\sim k; t \rangle
\]
with the analogies \( R(t) \rightarrow A(t) \) and \( | \alpha; t \rangle \rightarrow | n\sim k; t \rangle \). Due to minimal coupling (20), the eigenvalues are given by
\[
\epsilon_n(k; t) = \epsilon_n(k_t).
\]
The tilde on \( | n\sim k; t \rangle \) emphasizes the adiabatic time evolution from (7)/(11), [83]
\[
\partial_t | n\sim k; t \rangle = -q E(t) \sum_{n \neq n'} | n\sim k; t \rangle \langle n\sim k; t | \partial_A | n\sim k; t \rangle
\]
such that the condition of parallel transport (14) in the adiabatic basis \( | n\sim k; t \rangle \) is satisfied,
\[
E(t) \langle n\sim k; t | \partial_A | n\sim k; t \rangle = 0.
\]
We note that the matrix elements used for the time evolution (23) are
\[
\langle n\sim k; t | i \partial_t | n'\sim k; t \rangle = -E(t) q \langle n\sim k; t | i \partial_A | n'\sim k; t \rangle = E(t) \delta_{n' n} \langle k; t | n\sim k; t \rangle
\]
introducing the dipole matrix element
\[ \hat{d}_{nn'}(k; t) := -q \langle n k; t | \frac{\partial}{\partial A} | n' k; t \rangle , \] (26)
with diagonal elements
\[ \hat{A}_n(k; t) := -q \langle n k; t | \frac{\partial}{\partial A} | n k; t \rangle \] (27)
known as the Berry connection. We arrive at a compact notation for the condition of parallel transport,
\[ \mathbf{E}(t) \cdot \hat{A}_n(k; t) = 0 . \] (28)

C. EoM for the density matrix in adiabatic basis

In the adiabatic basis defined in (6) and (7), the EoM (3) takes the form (in the absence of collisions)
\[ i \langle \alpha; t | \rho | \beta; t \rangle = \tilde{\epsilon}_{\alpha\beta}(t) \rho_{\alpha\beta}(t) + i \langle \alpha | \rho | \tilde{\beta}; t \rangle \] (29)
where we define \( \tilde{\epsilon}_{\alpha\beta}(t) = \tilde{\epsilon}_\alpha(t) - \tilde{\epsilon}_\beta(t) \) and \( \rho_{\alpha\beta}(t) := \langle \alpha; t | \rho | \beta; t \rangle \).

To arrive at a closed set of equations for the matrix elements of \( \rho \) in the adiabatic frame, we need to reformulate (29) such that derivative of matrix elements of \( \rho \) appear rather than matrix elements of \( \rho \). To arrive at such an EoM for the matrix elements, we will employ the relation
\[ i \frac{d}{dt} \langle \alpha; t | \rho | \beta; t \rangle = \tilde{\epsilon}_{\alpha\beta}(t) \rho_{\alpha\beta} + i \langle \alpha | \rho | \tilde{\beta}; t \rangle \]
where (29) has been used; on the rhs the time variable has been suppressed and a short-hand notation \( \partial_i | \alpha; t \rangle = | \alpha; t \rangle \) was introduced. Inserting the resolution of the identity, \( \mathbb{I} = \sum_\alpha | \alpha \rangle \langle \alpha | \), we find
\[ \left( i \frac{d}{dt} - \tilde{\epsilon}_{\alpha\beta}(t) \right) \rho_{\alpha\beta} = i \sum_\alpha \langle \alpha | \tilde{\alpha} \rangle \rho_{\alpha\beta} + \rho_{\alpha\beta} \langle \alpha | \tilde{\beta} \rangle . \] (30)

With \( \langle \alpha | \tilde{\alpha} \rangle = -\langle \tilde{\alpha} | \alpha \rangle \) and Eq. (11), we conclude
\[ \left( i \frac{d}{dt} - \tilde{\epsilon}_{\alpha\beta}(t) \right) \rho_{\alpha\beta} = \mathbf{R}(t) \sum_\alpha \rho_{\alpha\beta} \langle \alpha | i \frac{\partial}{\partial \mathbf{R}} \rangle - \langle \alpha | i \frac{\partial}{\partial \mathbf{R}} \rangle \rho_{\alpha\beta} \]
arriving at the explicit form of the general EoM in the adiabatic frame.

Semiconductor Bloch equations. In the presence of a crystal symmetry (and in the absence of mean-field interactions) the equation of motion of the density operator, Eq. 4, takes a block-diagonal form
\[ i \dot{\rho}(k) = [h(k; t), \rho(k)] \] (32)
where each block has a common \( k \)-vector and, analogous to Eq. (17), \( h(k; t) \) and \( \rho(k) \) are matrices that act within the Hilbert space of the unit cell (“bands”). The matrices \( h(k; t) \) and \( \rho(k) \) are defined via their matrix elements:
\[ h_{nn'}(k; t) = \langle n k | h(t) | n' k \rangle = \langle n k | h(k; t) | n' k \rangle \] (33)
and similarly for \( \rho_{nn'}(k) \), see Appendix A for further details. Electric fields are readily treated in the Coulomb gauge: \( h(k; t) = h^{\text{in}}(k - \mathbf{A}(t)) \). The stationary basis used in (33) can be rotated into the adiabatic Bloch states from (15)-(27) with the analogies \( \mathbf{R}(t) \rightarrow \mathbf{A}(t) \) and \( | \alpha; t \rangle \rightarrow | n k; t \rangle \). The results of the previous section then translate into
\[ \left( i \frac{d}{dt} - \epsilon_{nn'}(k_i) \right) \tilde{\rho}_{nn'}(k; t) = \mathbf{R}(t) \sum_\alpha \tilde{\rho}_{nn'}(k; t) \mathbf{d}_{nn'}(k; t) - \mathbf{d}_{nn'}(k; t) \mathbf{d}_{nn'}(k; t) \] (34)
with the density matrix \( \tilde{\rho}_{nn'}(k; t) \) in the adiabatic basis and defining
\[ \epsilon_{nn'}(k_i) := \epsilon_n(k_i) - \epsilon_{n'}(k_i) \]
Eqs. (34) are known as the semiconductor Bloch equations (SBE). They have been derived here emphasizing a geometric perspective. Note that due to translational invariance, in (34) only diagonal matrix elements of \( \tilde{\rho} \) with a single \( k \)-point appear, see Appendix B for details. Another remarkable property of Eq. (34) is that matrix elements \( \tilde{\rho}_{nn'}(k; t) \) taken at different wavevectors \( k \) do not couple due to translational invariance of \( \mathbf{A}(t) \); terms involving gradients \( \partial_k \) are absent in (34), which otherwise appear; see Appendix B for further details.

D. Co-moving basis and EoM for its density matrix

We categorize the basis sets introduced before by considering a mapping \( f: \{ n, 1, \text{BZ}, \mathbb{R} \} \rightarrow \mathcal{H} \), where \( \mathcal{H} \) is the Hilbert space of Bloch states. We regard \( | n k; t \rangle \) as such a function \( f \) with variables \( n \), \( k \) and \( t \), that, when evaluated for a given \( n \), \( k \) and \( t \), returns a state in \( \mathcal{H} \). All of these functions are collected in the set
\[ F := \left\{ | n k; t \rangle : \{ n, 1, \text{BZ}, \mathbb{R} \} \rightarrow \mathcal{H} \right\} . \]

We define the set of instantaneous functions \( I \) containing every function \( | n k; t \rangle \) that is an eigenstate of \( h(k; t) \) for each instantaneous \( (k, t) \) pair,
\[ I := \left\{ | n k; t \rangle \in F : h(k; t) | n k; t \rangle \stackrel{(21)}{=} \epsilon_n(k; t) | n k; t \rangle \right\} . \]

Next, we define a subset of \( I \) that has the special property that the phase factors evolve smoothly in time, i.e., the functions are differentiable in time,
\[ D := \left\{ | n k; t \rangle \in I : | n k; t \rangle \text{ differentiable in } t \right\} . \]
In the same spirit, we define the *adiabatic* subset of functions that additionally fulfill the adiabatic time evolution (23),

\[
A := \left\{ |n\mathbf{k}; t\rangle \in D : \partial_t |n\mathbf{k}; t\rangle = -qE(t) \sum_{\alpha \neq n} \langle n\mathbf{k}; t| \langle n\mathbf{k}; t| \partial_\mathbf{A} |n\mathbf{k}; t\rangle \right\}.
\]

We further define the set \(S\) of stationary (i.e. time-independent), differentiable-in-k functions for a stationary basis \( |n\mathbf{k}\rangle \) to \( \{ \mathbb{B}, 1, \text{BZ}\} \rightarrow \mathcal{H} \),

\[
S := \left\{ |n\mathbf{k}\rangle : h^{\text{in}}(\mathbf{k})|n\mathbf{k}\rangle = \epsilon_n(\mathbf{k})|n\mathbf{k}\rangle \right\}.
\]

**Bloch electrons in homogeneous electric field.**

For the dynamics of Bloch electrons in a homogeneous electric field, we have \( h(\mathbf{k}; t) = h^{\text{in}}(\mathbf{k} - \mathbf{A}(t)) \). It is convenient to introduce a set of *co-moving* functions as

\[
C := \left\{ |n\mathbf{k}; t\rangle \in D : \text{there is a } |n\mathbf{k}\rangle \in S \right\},
\]

such that \( |n\mathbf{k}; t\rangle = |n\mathbf{k}\rangle \),

using the definition \( \mathbf{k}_t := \mathbf{k} - \mathbf{A}(t) \) from (20). The co-moving set forms a basis that is also known as *Houston basis* [84] in the literature. We mention that a co-moving function \( |n\mathbf{k}; t\rangle \in C \) is an eigenstate of \( h(\mathbf{k}; t) \) with eigenvalue \( \epsilon_n(\mathbf{k}_t) \), see (16). In general, a co-moving function is not adiabatic,

\[
C \not\subseteq A,
\]

that means, the condition of parallel transport, Eq. (28), is violated by a general co-moving function. The only degree of freedom that distinguishes an adiabatic function \( |n\mathbf{k}; t\rangle \in A \) and a co-moving function \( |n\mathbf{k}_t\rangle \) is a differentiable phase [78] \( \gamma_n(\mathbf{k}_t, t) \) such that

\[
|n\mathbf{k}; t\rangle = \exp(i \gamma_n(\mathbf{k}_t, t)) |n\mathbf{k}_t\rangle.
\]

The dipole moment and the Berry connection from (26) and (27) when expressed in the co-moving basis (35), \( |n\mathbf{k}; t\rangle = |n\mathbf{k}_t\rangle \), turn into familiar expressions [18–20, 79, 85]

\[
d_{nn'}(\mathbf{k}_t) = q \langle n\mathbf{k}_t| \partial_\mathbf{k}_t |n'\mathbf{k}_t\rangle,
\]

\[
\mathcal{A}_n(\mathbf{k}_t) = q \langle n\mathbf{k}_t| \partial_\mathbf{k}_t |n\mathbf{k}_t\rangle.
\]

For deriving an equation of motion for the density matrix in the co-moving basis \( |n\mathbf{k}_t\rangle \), we proceed similarly as for deriving Eq. (34): In Eq. (31), the substitutions \( \mathbf{R}(t) \rightarrow \mathbf{A}(t) \) and \( |\alpha; t\rangle \rightarrow |n\mathbf{k}_t\rangle \) lead to the familiar form of the SBE in the co-moving basis as [18–20, 85]

\[
\left( i \frac{d}{dt} - \epsilon_{nn'}(\mathbf{k}_t) \right) \varrho_{nn'}(\mathbf{k}_t) = E(t) \sum_{\alpha} \varrho_{n\alpha}(\mathbf{k}_t) d_{\alpha n'}(\mathbf{k}_t) - d_{n\alpha}(\mathbf{k}_t) \varrho_{n'\alpha}(\mathbf{k}_t).
\]

The co-moving basis is our preferred basis for numerical calculations since dipoles and Berry connections, (37) and (38), are easy to compute.

**E. Gauge perspective of the EoM**

So far, we have derived equations of motion for density matrices, with examples focusing on homogeneous electric fields treated in Coulomb-gauge with \( \mathbf{E} = -\mathbf{A} \). Then, the operator relation (32) takes the form

\[
i \partial_t \rho(\mathbf{k}; t) = [h^{\text{in}}(\mathbf{k} - \mathbf{A}(t)), \rho(\mathbf{k}; t)].
\]

In this section we translate this commutator relation into an EoM for matrix elements of \( \rho(\mathbf{k}; t) \). We represent \( \rho(\mathbf{k}; t) \) in two different basis sets and present the EoM associated with either one.

Within the co-moving basis \( |n\mathbf{k}_t\rangle \), we have matrix elements \( \varrho_{nn'}(\mathbf{k}; t) \) and

\[
\varrho_{nn'}(\mathbf{k}; t) := \langle n\mathbf{k}_t| \rho(\mathbf{k}; t) |n'\mathbf{k}_t\rangle,
\]

see Appendix C where we show that Eqs. (40) and (41) indeed lead to the EoM (39).

For exploring another basis, we define a boost operator as

\[
\mathfrak{B}(t) := \Xi e^{-\int dt' \mathbf{k} \partial_{k_t}}
\]

where the operator \( \Xi \) keeps track of the proper ordering along the \( \mathbf{k} \)-space trajectory; by definition, it acts on stationary Bloch states as

\[
\mathfrak{B}(t) |n\mathbf{k}\rangle = |n\mathbf{k}_t\rangle.
\]

For the case of a homogeneous electric field, we have \( \mathbf{k}_t = \mathbf{k} - \mathbf{A}(t) \) such that functions are shifted as \( \mathfrak{B}(t) f(\mathbf{k}) = f(\mathbf{k} - \mathbf{A}(t)) \) (see Appendix D) in line with Eq. (43). One may interpret the boost operator as analogous to the generator of translation that is a function of the momentum operator. By applying the boost operator

\[
h^{\text{in}}(\mathbf{k} - \mathbf{A}(t)) = \mathfrak{B}(t) h^{\text{in}}(\mathbf{k}) \mathfrak{B}^{-1}(t)
\]

we translate the initial, unperturbed Hamiltonian \( h^{\text{in}}(\mathbf{k}) \) to the time-dependent Hamiltonian \( h(\mathbf{k}; t) \) at time \( t \).

With the definition of the density matrix in the dipole gauge [86]

\[
\rho^D(\mathbf{k}; t) := \mathfrak{B}^{-1}(t) \rho(\mathbf{k}; t) \mathfrak{B}(t)
\]

one can derive an EoM from Eq. (40) as

\[
i [\partial_t + \mathbf{k}(t) \partial_\mathbf{k}_t] \rho^D(\mathbf{k}; t) = [h^{\text{in}}(\mathbf{k}), \rho^D(\mathbf{k}; t)].
\]

In this representation, the commutator involves the unperturbed Hamiltonian, only. It therefore is evaluated conveniently in the stationary basis \( |n\mathbf{k}\rangle \). Similarly to
Appendix C, one derives the traditional dipole-gauge formulation of the SBE with the characteristic gradient term on the lhs. [33]

\[ i[\partial_t + qE(t)\partial_k]\rho_{nn'}(k; t) = \epsilon_{nn'}(k)\rho_{nn'}^D(k; t) + E(t)\sum_{n'} (\rho_{nn'}^D(k; t)d_{nn'}(k) - d_{nn}(k)\rho_{nn'}^D(k; t)). \] (47)

using the definition

\[ \rho_{nn'}^D(k; t) := \langle nk|\rho^D(k; t)|nk\rangle. \] (48)

As shown in Appendix D, this definition relates to the Coulomb-gauge density matrix elements in the co-moving basis from Eq. (41) via

\[ \varrho_{nn'}(k; t) = \rho_{nn'}^D(k - A(t); t). \] (49)

An alternative way to derive Eq. (47) starts from the dipole (or length) gauge in which the electric field is represented by a linear potential. The relation Eq. (45) between \( \rho(k) \) and \( \rho^D(k) \) is thus understood as a gauge transformation. We emphasize that the time evolution of physical observables resulting from the SBE is gauge-independent, of course. [85, 87]

### III. OBSERVABLES: EMISSION INTENSITY, DYNAMICAL POLARIZATION AND CURRENT

As a response to the time-dependent perturbing fields, the charge density is accelerated; it varies in time and therefore irradiates light. The calculation of the emitted light intensity starts from the familiar equivalence between longitudinal current density and the derivative of the polarization. [20, 88, 89]

\[ j(t) = \partial_tP(t). \] (50)

Experiments measure the frequency resolved emission intensity \( I(\omega) \), which is given by [89]

\[ I(\omega) = \frac{\omega^2}{3\pi^3} |j(\omega)|^2. \] (51)

In the following, we derive expressions for the (dynamical) polarization and the current of the emitted radiation.

#### A. Dynamical polarization \( P \)

We compute the polarization [90] as expectation value of the dipole operator \( qr \) in a general basis \( |\alpha\rangle \) from (1) as [20]

\[ P(t) = \frac{1}{V} \text{Tr} \langle qr|\rho(t)\rangle = \frac{1}{V} \sum_{\alpha,\beta} \langle \alpha|qr|\beta\rangle \rho_{\beta\alpha}(t) \]

\[ = \frac{1}{V} \sum_{mm'} \sum_{kk'} \langle mk|qr|m'k'\rangle \rho_{mm'}(k'k; t). \] (52)

with \( V \) denoting the normalization volume. Adopting the notation from Eq. (15), we employ a basis \( |mk\rangle \) with \( k \)-independent lattice-periodic part \( |m\rangle \),

\[ \langle r|mk\rangle = \frac{1}{\sqrt{V}} e^{ikr} \langle r|m\rangle. \] (53)

The major advantage of the \( |m\rangle \)-basis over a \( k \)-dependent lattice-periodic part \( |nk\rangle \) is that gradient-terms in \( k \) can be much easier handled. We also derive our main result for \( \partial_tP \) using a \( k \)-dependent lattice-periodic \( |nk\rangle \) basis in Appendix E. We keep the full \( k \)-dependence of

\[ \rho_{mm'}(kk'; t) = \langle mk|\rho(t)|m'k'\rangle \]

in (52) to properly account for \( k \)-derivatives later on.

We evaluate the dipole matrix element \( \langle mk|r|m'k'\rangle \) appearing in the polarization (52) adopting (B1) as

\[ \langle mk|r|m'k'\rangle = \frac{(2\pi)^d}{V} \langle m|[i\partial_k e^{-i(k-k')r}\delta(k-k')]|m'\rangle. \] (54)

With (54) and results from Appendix A, we obtain

\[ P(t) = q \sum_{mm' BZ} \int_{BZ} dk \int_{BZ} dk' \frac{d}{(2\pi)^d} \rho_{mm'}(k'k; t) \]

\[ \times \langle m| \left[ i\partial_k e^{-i(k-k')r}\delta(k-k') \right] |m'\rangle. \]

\[ = iq \sum_{mm' BZ} \int_{BZ} dk \int_{BZ} dk' \frac{d}{(2\pi)^d} \delta_{mm'}(k'k; t) \]

\[ \times \frac{\partial \rho_{mm'}(k'k; t)}{\partial k'} \bigg|_{k'\to k} \]

\[ = iq \int_{BZ} \frac{dk}{(2\pi)^d} \text{Tr}_n \rho(k; t), \] (55)

where integration by parts has been used to arrive at the second equation. In the last line, we defined

\[ \rho_{mm'}(kk; t) := \langle \epsilon_m^\dagger \partial_k \epsilon_m \rangle = \frac{\partial \rho_{mm'}(k'k; t)}{\partial k'} \bigg|_{k'\to k}. \] (56)

The trace in Eq. (55) can be evaluated in any lattice periodic basis and it is our preferred choice to continue with basis-independent representations.

For computing the emission from Eq. (51), we employ the time derivative of \( P \) that translates to the time derivative of \( \rho \) in Eq. (55). We insert EoM (40), \( i\dot{\rho}(k; t) = [h(k; t), \rho(k; t)] \) in the Coulomb gauge in the right-hand side of (56) and obtain

\[ i\text{Tr}_n \dot{\rho}(k; t) = \text{Tr}_n \left[ [h(k; t), \rho(k; t)] + (\partial_k h(k; t)) \rho(k; t) \right]. \] (57)

Since the trace of the commutator vanishes, we have

\[ \partial_tP(t) = q \int_{BZ} \frac{dk}{(2\pi)^d} \text{Tr}_n \left[ \partial_k h(k; t) \right] \rho(k; t). \] (58)
As an application, we evaluate the trace in (58) for the special case of a homogeneous electric field, \( h(\mathbf{k}; t) = h_{\text{in}}(\mathbf{k} - \mathbf{A}(t)) \) in the co-moving basis \(|n\mathbf{k}; t\rangle = |n\mathbf{k}_i\rangle\):

\[
\partial_t \mathbf{P}(t) = q \sum_{nn'} \int_{BZ} \frac{d\mathbf{k}}{(2\pi)^d} \langle n\mathbf{k}; t | \frac{\partial h_{\text{in}}(\mathbf{k} - \mathbf{A}(t))}{\partial \mathbf{k}} | n'\mathbf{k}; t \rangle \partial_{n'\mathbf{k}}(\mathbf{k}; t) \tag{59}
\]

so that the density matrix \( \partial_{n'\mathbf{k}}(\mathbf{k}; t) \) as defined in Eq. (41) in the co-moving basis appears. In this way, it is possible to use \( \partial_{n'\mathbf{k}}(\mathbf{k}; t) \) from the dynamics in Eq. (39) to evaluate \( \partial_t \mathbf{P}(t) \) and subsequently also the emission intensity.

The transparent result (59) implies that the velocity associated with the co-moving states \(|n\mathbf{k}; t\rangle \) as given by the matrix element derives from the instantaneous band structure. Notice, however, that this particular aspect of (59) is a consequence of our choice of gauge. In the later Section III C an equivalent expression, Eq. (64), will be derived for the current density that involves the unperturbed band-structure.

### B. Longitudinal current density \( \mathbf{j} \)

An alternative derivation of (58) embarks on the relation (50) between the longitudinal charge current density and the polarization, \( \mathbf{j}(t) = \mathbf{P} \) and

\[
\mathbf{j}(t) = \frac{1}{V} \text{Tr}[\mathbf{q} \rho(t)] = \frac{1}{V} \sum_{\alpha,\beta} \langle \alpha | \mathbf{q} | \beta \rangle \rho_{\beta\alpha}(t)
\]

\[
= \mathcal{V} \sum_{nn'} \int_{BZ} \frac{d\mathbf{k}}{(2\pi)^d} \frac{d\mathbf{k}'}{(2\pi)^d} \langle n\mathbf{k} | \mathbf{q} | n'\mathbf{k}' \rangle \rho_{nn'}(n'\mathbf{k}; t) . 
\tag{60}
\]

Since the velocity operator \( \dot{\mathbf{r}} \) relates to the Hamiltonian via the operator derivative \( \dot{\mathbf{r}} = \partial h/\partial \mathbf{p} \), we readily conclude

\[
\mathbf{j}(t) = q \mathcal{V} \sum_{nn'} \int_{BZ} \frac{d\mathbf{k}}{(2\pi)^d} \frac{d\mathbf{k}'}{(2\pi)^d} \langle n\mathbf{k} | \ dozen(t) \dot{\mathbf{r}} | n'\mathbf{k}' \rangle \rho_{nn'}(n'\mathbf{k}; t) . 
\tag{61}
\]

**Translational invariance:** In the special situation of translational invariance, \( h \) is diagonal in the eigenstates \(|n\mathbf{k}\rangle\) of the momentum operator \( \mathbf{p} \). Therefore, first the operator derivative \( \partial/\partial \mathbf{p} \) in Eq. (61) can be replaced by \( \partial/\partial \mathbf{k} \) and second, the matrix element is proportional to \( \delta(\mathbf{k} - \mathbf{k}') \). [78] Hence, Eq. (61) simplifies to

\[
\mathbf{j}(t) = q \mathcal{V} \int_{BZ} \frac{d\mathbf{k}}{(2\pi)^d} \text{Tr}_{n} \left[ \frac{\partial h(\mathbf{k}; t)}{\partial \mathbf{k}} \rho(\mathbf{k}; t) \right] . \tag{62}
\]

and we recover (58).

### C. Relation to earlier work

We line out the relation of our results (59) to frequently cited formula from the literature [33, 34] that distinguish intra- and interband contributions to the current and the emission. In these works the density matrix \( \rho^D \), i.e. in the dipole gauge, is used. For convenient comparison we express the current density in terms of \( \rho^D \). To this end, we embark on the trace formula Eq. (58)

\[
\mathbf{j}(t) = q \int_{BZ} \frac{d\mathbf{k}}{(2\pi)^d} \text{Tr}_{n} \left[ \partial_{n'\mathbf{k}} h_{\text{intr}}(\mathbf{k}; t) \rho^D(\mathbf{k}; t) \right] .
\]

Recalling Eq. (45) and (44), we derive an expression in the dipole gauge as

\[
\mathbf{j}(t) = q \int_{BZ} \frac{d\mathbf{k}}{(2\pi)^d} \text{Tr}_{n} \left[ \mathfrak{A}_{n}^{-1} \partial_{n'\mathbf{k}} h_{\text{intr}}(\mathbf{k}; \mathbf{A}(t)) \mathfrak{B} \rho^D(\mathbf{k}; t) \right]
\]

\[
= q \int_{BZ} \frac{d\mathbf{k}}{(2\pi)^d} \text{Tr}_{n} \left[ \mathfrak{A}_{n}^{-1} \partial_{n'\mathbf{k}} \mathfrak{B} h_{\text{intr}}(\mathbf{k}) \rho^D(\mathbf{k}; t) \right]
\]

\[
= q \int_{BZ} \frac{d\mathbf{k}}{(2\pi)^d} \text{Tr}_{n} \left[ \partial_{n'\mathbf{k}} h_{\text{intr}}(\mathbf{k}) \rho^D(\mathbf{k}; t) \right] 
\tag{63}
\]

where the last line is assuming \( \mathbf{k} \) does not depend on \( \mathbf{k} \), as is the case for homogeneous electric fields. The trace in Eq. (63) when evaluated in the stationary basis \(|n\mathbf{k}\rangle\) reads

\[
\mathbf{j}(t) = q \int_{BZ} \frac{d\mathbf{k}}{(2\pi)^d} \text{Tr}_{n} \left[ \langle n\mathbf{k} | \partial_{n'\mathbf{k}} h_{\text{intr}}(\mathbf{k}) | n'\mathbf{k}' \rangle \rho^D_{nn'}(\mathbf{k}; t) \right] . 
\tag{64}
\]

In Appendix F we derive an expression for the matrix element

\[
\langle n\mathbf{k} | \partial_{n'\mathbf{k}} h_{\text{intr}}(\mathbf{k}) | n'\mathbf{k}' \rangle = \delta_{nn'} \partial_{n'\mathbf{k}} \epsilon_{n}(\mathbf{k}) - \frac{i}{q} \text{d}_{nn'}(\mathbf{k}) \epsilon_{n'\mathbf{k}}(\mathbf{k}) \tag{65}
\]

that can motivate the traditional splitting of (64) into intraband (\( n=n' \)) contributions and a rest (\( n \neq n' \)). For the intraband current we reproduce the familiar expression, [33, 34]

\[
\mathbf{j}_{\text{intr}}(t) := q \int_{n} \frac{d\mathbf{k}}{(2\pi)^d} \partial_{n} \epsilon_{n}(\mathbf{k}) \rho^D_{nn}(\mathbf{k}; t) \tag{66}
\]

while for the remainder, \( \delta \mathbf{j} := \mathbf{j} - \mathbf{j}_{\text{intr}} \), we obtain

\[
\delta \mathbf{j}(t) = -i \int_{n \neq n'} \frac{d\mathbf{k}}{(2\pi)^d} \text{d}_{nn'}(\mathbf{k}) \epsilon_{n'\mathbf{k}}(\mathbf{k}) \rho^D_{n'n}(\mathbf{k}; t) \tag{67}
\]
where the diagonal terms vanish since \( \epsilon_{nn}(k) = 0 \). Embarking on (47) we can also write

\[
\delta j(t) = \sum_{n \neq n'} \int \frac{dk}{(2\pi)^d} \mathbf{d}_{nn'}(k) \left( \partial_t + qE(t) \partial_k \right) \rho^D_{nn'}(k; t) + \sum_{n \neq n'} \int \frac{dk}{(2\pi)^d} \mathbf{d}_{nn'}(k) \left[ E(t) \mathbf{d}(k), \rho^D(k; t) \right]_{nn'} . \tag{68}
\]

The first term in (68)

\[
\partial_t \mathbf{P}^\text{inter}(t) := \sum_{n \neq n'} \int \frac{dk}{(2\pi)^d} \mathbf{d}_{nn'}(k) \dot{\rho}^D_{nn'}(k; t) \tag{69}
\]

reproduces the usual expression [33, 34] for the interband currents expressed in terms of the time derivative of the interband polarization \( \mathbf{P}^\text{inter} \). The remaining terms in (68) are captured introducing a (non-linear) time-dependent conductance tensor

\[
\sigma_{ij}(t) := q \sum_{n \neq n'} \int \frac{dk}{(2\pi)^d} d^{(i)}_{nn'}(k) \left( \partial_k \rho^D_{nn'}(k; t) + \left[ d^{(j)}(k), \rho^D(k; t) \right]_{nn'} \right) = q \sum_{n \neq n'} \int \frac{dk}{(2\pi)^d} \left( - (\partial_k d^{(i)}_{nn'}(k)) \rho^D_{nn'}(k; t) + d^{(i)}_{nn'}(k) d^{(j)}(k), \rho^D(k; t) \right) \tag{70}
\]

where \( d^{(i)} \) and \( \partial_k \) denote the components \( i \) and \( j \) of the corresponding vector-valued objects \( \mathbf{d} \) and \( \partial_k \). Collecting terms, we have

\[
j(t) = \partial_t \mathbf{P}^\text{inter}(t) + j^\text{intra}(t) + \sigma(t) \mathbf{E}(t) . \tag{71}
\]

To the best of our knowledge this relation has been derived here for the first time; in particular, the term \( \sigma(t) \mathbf{E}(t) \) was not included in previous theoretical treatises. As we will show in Sec. IV C, its contribution to high-harmonic generation can be sizable for Dirac-type systems.

In the literature, the third term in (71) has not been accounted for. [33, 34] Therefore, it is important to note that \( \sigma(t) \) vanishes when the following two conditions on \( \mathbf{d}_{nn'}(k) \) are satisfied: all diagonal entries vanish, \( \mathbf{d}_{nn} = 0 \) and the off-diagonals point in the same direction, \( \mathbf{d}_{nn'}(k) = \mathbf{d}_{nn'} \hat{e} \), with \( \mathbf{d}_{nn'} \) independent of \( k \). Both conditions are apparent since the second term in Eq. (70) contributes with

\[
\sum_{n \neq n', n} d^{(i)}_{nn'} \left[ d^{(j)}_{nn'}, \rho^D_{nn'} - \rho^D_{nn'} d^{(j)}_{nn'} \right] = \text{Tr} \left[ \left[ d^{(i)}, d^{(j)} \right] \rho^D \right] - \sum_n d^{(i)}_{nn} d^{(j)}_{nn}, \rho^D_{nn} .
\]

Indeed, the corresponding simplified form of \( \mathbf{d}_{nn'}(k) \) has frequently been adopted [33, 34], such that \( \sigma(t) \) vanishes and the corresponding results remain unaffected.

IV. APPLICATION: DYNAMICS OF DIRAC FERMIONS

Motivated by recent experiments [91–96], we apply the SBE formalism to the density matrix dynamics for a Dirac-type dispersion driven by an ultra-short electric field pulse. We here focus on bandstructure effects and thus neglect mean-field interactions.

A. The model

Hamiltonian. We employ a two-dimensional Dirac cone

\[
h^\text{in}(k) = v_F (k_y \sigma_x - k_x \sigma_y) \tag{72}
\]

with a Fermi velocity \( v_F = 4.3 \times 10^5 \text{ m/s} = 1.44 \times 10^{-3} \text{ c} \) that is a prototypical two-band surface Hamiltonian of a topological insulator as bismuth telluride (Bi\textsubscript{2}Te\textsubscript{3}). [97] Such a model Hamiltonian can be obtained, e.g., from \textit{ab-initio} calculations by \( k \cdot p \) perturbation theory [97] or the use of Wannier functions [98, 99]. The eigenstates and bandstructure are computed as

\[
|\mathbf{n}k\rangle = \frac{1}{\sqrt{2}} \left( -1 + 2 \delta_{\mathbf{nv}} \right) , \quad \epsilon_n(k) = v_F |k| (1 - 2 \delta_{\mathbf{nv}}) , \tag{73}
\]

and the dipoles follow

\[
\mathbf{d}_{nn'}(k) = - \frac{\hat{e}_\theta}{2 |k|} \tag{74}
\]

for all \( n, n' \in \{v, c\} \) being the valence or conduction band, with \( \theta \) being the polar angle and \( \hat{e}_\theta \) the unit vector orthogonal to \( k \).

Electric-field pulse. An ultra-short laser pulse is employed with an electric driving field that is polarized in \( x \)-direction,

\[
\mathbf{E}(t) = E \hat{e}_x \sin(\omega_0 t) \exp \left( - \frac{t^2}{\sigma^2} \right) , \tag{75}
\]

where \( \omega_0 = 2 \pi \times 25 \text{ THz}, \quad E = 5 \text{ MV/cm} \) and \( \sigma = \omega_0 / (4 \pi) \) throughout our calculations. The pulse shape here adopted follows the experimental ones. [2, 4, 100]

Equations of motion. The EoM will be adopted from Eq. (39). In order to mimic the effect of the collision term \( \partial \rho / \partial t \rvert_{\text{coll}} = 0 \) we include it phenomenologically by adding a damping of offdiagonal density matrix elements [20, 63]:

\[
\left( i \frac{\partial}{\partial t} + i \frac{(1 - \delta_{nn'})}{T_2} - \epsilon_{nn'}(k_i) \right) \varrho_{nn'}(k; t) = \mathbf{E}(t) \sum_{n} \varrho_{nn}(k; t) \mathbf{d}_{nn'}(k_i) - \mathbf{d}_{nn}(k_i) \varrho_{nn'}(k; t) , \tag{76}
\]

8
where \( \mathbf{k} = \mathbf{k} - \mathbf{A}(t) \), \( \dot{\mathbf{A}}(t) = -q\mathbf{E}(t) \). For practical calculations, we have chosen \( T_2 = 1 \) fs following Ref. 4, similar to Refs. 12, 23, and 101; for further discussion see Ref. 63.

The initial condition for integrating the EoM was chosen with the valence band being filled and the conduction band being empty:

\[
\varrho_{n\nu}(\mathbf{k}) = \delta_{n\nu}\delta_{n'\nu}.
\]  

(77)

For the \( k \)-domain of integration, we have allowed for the limit \( \pi/a \to \infty \). This corresponds to a situation where the linear size of the Brillouin zone \( \pi/a \) exceeds any other characteristic wavenumber; in particular, \( \pi/a \gg \omega_0/v_F \) and \( \pi/a \gg qE/\omega_0 \). The inequalities are satisfied for typical lattice constants \( a \) and parameters \( \omega_0, v_F, E \) as chosen in this work: For \( a = 3 \) Å, we have \( \pi/a \approx 2qE/\omega_0 \) such that Bloch electrons excited at the \( \Gamma \)-point hardly touch the Brillouin zone boundary. Much higher field strengths up to 72 MV/cm are used to drive the Bloch electrons over the Brillouin zone boundary to initiate Bloch oscillations [2].

Finally, the emission intensity \( I(\omega) \) is computed from Eq. (51) using the longitudinal current density, Eq. (59),

\[
\bar{j}(t) = q \sum_{n\nu} \int \frac{d\mathbf{k}}{(2\pi)^d} \langle n \mathbf{k} | \frac{\partial \varrho_{n\nu}(\mathbf{k})}{\partial \mathbf{k}} | n' \mathbf{k} \rangle \varrho_{n'\nu}(\mathbf{k}) \pi t.
\]  

(78)

B. The method

For integrating the EoM in Eq. (76), we use a backward differentiation formula with a maximum adaptive timestep of 0.1 fs as implemented in scipy [102].

Convergence tests. We investigate the convergence of the \( k \)-point integration of Eq. (78) in Fig. 1. As \( k \)-point mesh, we choose a \( \Gamma \)-centered Monkhorst-Pack mesh [103] that is confined by a rectangle. As shown in Fig. 1 (a), the emission intensity converges when increasing the size of the \( k \)-mesh and the density of \( k \)-points in direction of the \( E \)-field (\( x \)-direction). Convergence is found for 1200 \( k \)-points and a length 1500 \( \omega_0/v_F \) in \( k_x \)-direction.

From Fig. 1 (b), we observe that the emission intensity converges when increasing the length and density of the \( k \)-mesh orthogonal to the electric driving field (\( k_y \)-direction). Here, convergence is found for 100 \( k \)-points and a length of 240 \( \omega_0/v_F \) in \( k_y \)-direction. We are left to choose a rectangular 1200 \( \times \) 100 \( k \)-mesh with size 1500 \( \times \) 240 \( \omega_0/v_F \) for all following \( k \)-integrations.

C. Results: Dynamics in homogeneous \( E \)-field

In Fig. 1, we find fast exponential decay by four orders of magnitude from the fundamental peak to fifth harmonic order (\( \omega = 5 \omega_0 \)). The decay for subsequent orders up to 20th harmonic order is slower. Similar behaviour has been observed in the literature for dynamics and emission from a semimetallic Hamiltonian [26] and the Haldane model [15].

Approximate dynamics. We continue our discussion by computing the emission intensity from the frequently cited result [33, 34] that ignores the term \( \sigma(t)\mathbf{E}(t) \) in Eq. (71),

\[
\bar{j}(t) \approx \bar{j}^{\text{intra}}(t) + \partial_t \mathbf{P}^{\text{inter}}(t).
\]  

(79)

We obtain the density matrix \( \rho^\text{D}(\mathbf{k}; t) \) in the stationary basis with the dipole gauge from the dynamics in Eq. (47) including off-diagonal damping \( T_2 \),

\[
\i \left[ \frac{\partial}{\partial t} + \frac{1 - \delta_{nn'}}{T_2} + q\mathbf{E}(t) \cdot \frac{\partial}{\partial \mathbf{k}} \right] \rho_{nn'}^\text{D}(\mathbf{k}; t) = \epsilon_{nn'}(\mathbf{k}) \rho_{nn'}^\text{D}(\mathbf{k}; t) + \mathbf{E}(t) \sum_n \left( \rho_{nn'}^\text{D}(\mathbf{k}; t) \mathbf{d}_{nn'}(\mathbf{k}) - \mathbf{d}_{nn}(\mathbf{k}) \rho_{nn'}^\text{D}(\mathbf{k}; t) \right). \]  

(80)
As an extra numerical test proving the equivalence of gauges, we also compute the emission in dipole gauge from the exact expression Eq. (64) using the result of Eq. (80) as an input.

Fig. 2 displays our results. Two emission curves computed from the exact current are shown for Coulomb and dipole gauge that lie on top of each other demonstrating the expected equivalence of gauges. The emission computed from the approximate expression (79) is also shown (blue color) and we observe a discrepancy between the exact result that includes the term \( \sigma(t)E(t) \) and the approximate treatment from Eq. (79) that neglects the term \( \sigma(t)E(t) \). While the emission from the approximate and exact current qualitatively agree, quantitative discrepancies appear at high harmonics reaching an order of magnitude deviation at fifth harmonic order. We trace this back to the fact that the intraband current dominates for the fundamental and the third harmonic peak, see the orange line in Fig. 2. At high harmonics the interband-currents become relatively more important and those get sizable contributions from the term \( \sigma(t)E(t) \) missing in (79) when dipole-matrix elements have a significant dependency on \( k \), as is the case of the Dirac-system at hand.

**V. CONCLUSIONS AND OUTLOOK**

A derivation of the semiconductor Bloch equations (SBE) for the time evolution of the density matrix has been presented emphasizing the close relation to the Berry connection. This particular approach has the appealing feature that it lends itself to a semiclassical perspective on the SBE allowing for a simplified treatment of magnetic-field effects by Lorentz forces that will be presented in a forthcoming publication.

Also, expressions have been rederived connecting the density matrix to physical observables, specifically, to the current density. In addition to the traditional current, summing intraband and interband-polarization contributions \([33, 34]\), we have identified an extra term; it becomes sizable in situations where dipole-matrix elements depend strongly on the wavenumber. We have implemented an SBE solver and applied it to Dirac metals, motivated by the observation that dipoles are strongly \( k \)-dependent for Dirac fermions. We find that the extra term gives a significant contribution to the total current, in particular, to the high-harmonic generation: the emission intensity can deviate by more than an order of magnitude upon neglecting the extra term.

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**Appendix A: Basics of lattice periodicity**

We define \( \mathcal{V} \) as a volume containing several unit cells with lattice vectors \( \mathbf{R} \). Then, an integration over \( \mathcal{V} \) is given by integrating over individual cells,

\[
\int \mathcal{V} \, df(r) = \sum_\mathbf{R} \int_\mathbf{C} \, df(r + \mathbf{R}). \tag{A1}
\]

\( \mathbf{C} \) denotes the integration over the (primitive) unit cell.

For vectors \( \mathbf{k} - \mathbf{k}' \) from the first Brillouin zone, we recall

\[
\sum_\mathbf{R} e^{i(\mathbf{k} - \mathbf{k}')\mathbf{R}} = \mathcal{N} \delta_{\mathbf{k} \mathbf{k}'} \simeq \frac{(2\pi)^d}{\mathcal{V}_c} \delta(\mathbf{k} - \mathbf{k}') \tag{A2}
\]

where \( \mathcal{N} := \sum_\mathbf{R} \) denotes the number of unit cells in \( \mathcal{V} \) and \( \mathcal{V}_c = \mathcal{V}/\mathcal{N} \) is the volume of a unit cell. The right-hand side of (A2) in the limit of large \( \mathcal{N} \) implies

\[
\sum_\mathbf{k} f(\mathbf{k}) \simeq \frac{\mathcal{V}}{(2\pi)^d} \int_{\mathbb{BZ}} d\mathbf{k} \ f(\mathbf{k}), \tag{A3}
\]

where we integrate over the first Brillouin zone.

The eigenstates of the stationary, lattice-periodic Hamiltonian are Bloch-states \( \langle n | \mathbf{k} \rangle \). In the context of (15), we have defined the lattice periodic wavefunction as \( u_{n\mathbf{k}} \):

\[
\langle \mathbf{r} | n \mathbf{k} \rangle = \frac{1}{\sqrt{\mathcal{N}}} e^{i\mathbf{k}\mathbf{r}} \langle \mathbf{r} | n \mathbf{k} \rangle =: \frac{1}{\sqrt{\mathcal{N}}} e^{i\mathbf{k}\mathbf{r}} u_{n\mathbf{k}}(\mathbf{r}). \tag{A4}
\]
The double angular brackets indicate that the normalization volume for \( u_{nk}(r) \) is the unit cell \( \mathcal{C} \):
\[
\langle nk|f(r)|n'k' \rangle := \int_{\mathcal{C}} \frac{dV}{V} u_{nk}^*(r) u_{n'k}(r) = \delta_{nn'}, \tag{A5}
\]
while the normalization volume for Bloch states \( |nk\rangle \) is \( V \):
\[
\langle nk|n'k \rangle := \frac{1}{N} \int_{V} dr \; u_{nk}^*(r) u_{n'k}(r) = \delta_{nn'}. \tag{A6}
\]
This notation is also used to define an integration of lattice periodic functions over a single unit cell as
\[
\langle nk|f(r)|n'k' \rangle := \int_{\mathcal{C}} \frac{dV}{V} u_{nk}^*(r) f(r) u_{n'k}(r). \tag{A7}
\]
In contrast, for expectation values of Bloch states \( |nk\rangle \) we integrate over the whole volume \( V \) with a normalization \( 1/N \) stemming from \( (A4) \)
\[
\langle nk|f(r)|n'k' \rangle \overset{(A4)}{=} \frac{1}{N} \int_{V} \frac{dV}{V} u_{nk}^*(r) e^{-i(k-k')r} f(r) u_{n'k}(r) \tag{A8}
\]
For infinitely extended systems, we have \( N \to \infty \).
In case we have an operator as the Hamiltonian \( \hat{h} \) or the density matrix \( \rho \), that are not diagonal in \( r \), we frequently evaluate matrix elements as follows:
\[
\langle nk|h(t)|n'k \rangle = \int_{\mathcal{C}} \frac{dV}{V} \frac{dV'}{V'} \langle nk|r\rangle \langle r|h(t)|r' \rangle \langle r'|n'k \rangle dr \cdot dr'.
\]
\[
= \frac{1}{N} \int_{\mathcal{C}} \frac{dV}{V} \frac{dV'}{V'} \int e^{-i(k-k')r} \langle nk|r\rangle \langle r|h(t)|r' \rangle e^{i(k-k')r'} u_{n'k}(r') dr' \tag{A9}
\]
\[
= \frac{1}{N} \int_{\mathcal{C}} \frac{dV}{V} \frac{dV'}{V'} \int e^{-i(k-k')r} \langle nk|r\rangle \langle r|\hat{h}(t)|r' \rangle e^{i(k-k')r'} u_{n'k}(r') dr' \tag{A9}
\]
\[
= \langle nk|h(t)|n'k \rangle \tag{A9}
\]
where we used in (\*), that \( u_{nk}(r) \) is lattice-periodic and in the first and last step that the real-space basis is complete. In step (\#), we have defined the operator \( \hat{h}(k; t) \) via its real-space matrix elements as
\[
\langle r'|\hat{h}(k; t)|r' \rangle = \frac{1}{N} \sum_{kk'} e^{-i(k-k')r} \langle r + R|\hat{h}(t)|r' + R \rangle e^{i(k-k')R}. \tag{A10}
\]
Bloch states \( |nk\rangle \) are eigenstates of the initial, lattice-periodic, time-independent Hamiltonian \( \hat{h} \), addition to
\[
\hat{h} \equiv e_n(k) |nk\rangle \tag{A11}
\]
We have for the lattice periodic part \( |nk\rangle \) where we
\[
\langle nk|\delta_{nn'}|nk \rangle = \langle nk|\hat{h}|nk \rangle \overset{(A9)}{=} \langle nk|\hat{h}|nk \rangle \tag{A12}
\]
and after using the completeness \( 1 = \sum_n |nk\rangle \langle nk| \) we find the eigenvalue equation for the lattice periodic part
\[
\hat{h}|nk\rangle = e_n(k) |nk\rangle \tag{A13}
\]
that is used in Eq. (16).

**Appendix B: Matrix elements of local operators**

We derive an identity relating matrix elements of local operators \( f(r) \) in the basis \( |nk\rangle \) to matrix elements in the basis \( |nk\rangle \). Employing the basic definitions of periodicity from Appendix A, we have
\[
\langle nk|f(r)|n'k' \rangle \overset{(A4)}{=} \frac{1}{N} \int_{\mathcal{C}} \frac{dV}{V} \int_{\mathcal{C}} \frac{dV'}{V'} \langle nk|r\rangle \langle r|f(\mathbf{r})|r' \rangle \langle r'|n'k' \rangle dr \cdot dr'.
\]
\[
= \frac{1}{N} \int_{\mathcal{C}} \frac{dV}{V} \int_{\mathcal{C}} \frac{dV'}{V'} \int e^{-i(k-k')r} \langle nk|r\rangle \langle r|f(\mathbf{r})|r' \rangle e^{i(k-k')r'} \langle r'|n'k' \rangle dr \cdot dr'.
\]
\[
= \frac{1}{N} \int_{\mathcal{C}} \frac{dV}{V} \int_{\mathcal{C}} \frac{dV'}{V'} \int e^{-i(k-k')r} \langle nk|r\rangle \langle r|f(\mathbf{r})|r' \rangle e^{i(k-k')r'} \langle r'|n'k' \rangle dr \cdot dr'.
\]
\[
= \langle nk|f(\mathbf{r})|n'k' \rangle \tag{B1}
\]
Using identity (B1) we evaluate \( k \)-sums as follows:
\[
\int_{\mathcal{C}} \frac{dV'}{V} \langle nk|f(\mathbf{r})|n'k' \rangle \psi(\mathbf{k'}) = \frac{(2\pi)^d}{V} \int_{\mathcal{C}} \langle nk|f(\mathbf{r})|n'k' \rangle \psi(\mathbf{k'}) \tag{B2}
\]
As an application, we consider a Hamiltonian \( \hat{h}(t) \) with a vector potential that varies in time and space \( \mathbf{A}(r, t) \). The Schrödinger dynamics in Bloch-state representation reads
\[
i\partial_t \langle nk|\psi \rangle = \sum_{\mathbf{k}} \langle nk|h(t)|nk \rangle \langle nk|\psi \rangle \tag{B3}
\]
we arrive at
\[ \langle n| k | h(t) | n| k \rangle := \langle n| k | h(-i\nabla - A(r, t)) | n| k \rangle. \]

By virtue of (B2), rhs matrix element can be rewritten with the consequence that
\[ i\partial_t \langle n| k | \psi \rangle = \sum_{n} \left[ h(k-A(i\partial_{k'}, t)) \langle n| k | n'| k \rangle \langle n'| k | \psi \rangle \right]_{k'=k}. \quad (B4) \]

As is explicit from this result, the spatial dependency of \( A(r) \) mixes neighboring \( k \)-values as a manifestation of the broken translational invariance. For a homogeneous \( A \), however, \( k \)-coupling is absent, as one would expect.

**Appendix C: Density matrix in the Coulomb gauge in the co-moving basis**

We derive the expression (41) for the density matrix in the Coulomb gauge in the co-moving basis \( |n| k | t \rangle = |n| k \}, \quad \rho_{nn'}(k; t) := \langle n| k | \rho(k; t) | n'| k \rangle \]
starting from the dynamics (40) in the Coulomb gauge,
\[ i\partial_t \rho(k; t) = \left[ h^\text{in}(k-A(t)), \rho(k; t) \right], \]
that is projected on the co-moving basis \( |n| k \}),
\[ i\partial_t |n| k \rangle = \left[ h^\text{in}(k-A(t)), \rho(k; t) \right] |n| k \rangle \]
\[ = \langle n| k | h^\text{in}(k-A(t)) |n| k \rangle = -\epsilon_{nn'}(k)\langle n| k | \rho(k; t) | n'| k \rangle. \quad (C1) \]

We are interested in a time derivative of matrix elements instead of matrix elements of the time derivative of operators and therefore state
\[ \langle n| k | (\partial_t \rho(k; t)) | n'| k \rangle = \partial_t \langle n| k | \rho(k; t) | n'| k \rangle - \langle \partial_t | n| k \rangle | k \rangle | n'| k \rangle - \langle n| k | \rho(k; t) | \partial_t | n'| k \rangle. \]

With the resolution of the identity \( 1 = \sum_{n} | n| k \} \langle n| k |, \quad \partial_t | n| k \rangle = -\hat{A}(t) \partial_{k'} | n| k \rangle \quad (C1) \]
we arrive at
\[ \langle n| k | (\partial_t \rho(k; t)) | n'| k \rangle = \partial_t \langle n| k | \rho(k; t) | n'| k \rangle + q(E(t)) \sum_{n} \left[ \left( \partial_{k'} \langle n| k | n'| k \rangle \right) | n'| k \rangle - q(E(t)) \sum_{n} \left[ \langle n| k | \rho(k; t) | n'| k \rangle \right] | n'| k \rangle \quad (C2) \]

The dipole matrix elements (37)
\[ d_{nn'}(k_s) = q \langle n| k_s | k_s | n'| k_s \rangle \]

and proving Eq. (49).

The booster operator has been defined in the main text in Eq. (42) as
\[ \mathcal{B}(t) = \mathcal{E} e^{-\int dt' k \partial_k}. \]

We consider this operator as a successive, time-ordered infinitesimal shifting,
\[ \mathcal{B}(t) \approx \prod_{t'=\infty}^{t} \left( 1 + dt' \hat{A}(t') \right) \]
We use \( \hat{A}(t) = \partial_t (k-A(t)) = -\hat{A}(t) \) for a homogeneous electric field and Taylor expansion \( f(k-dt \hat{A}(t)) = (1-dt \partial_k \hat{A}(t)) f(k) \) to show
\[ \mathcal{B}(t) f(k) \approx \prod_{t'=\infty}^{t} \left( 1 - dt' \hat{A}(t') \right) \]

Next, we prove Eq. (49),
\[ \theta_{nn'}(k; t) = \rho_{nn'}(k-A(t); t) \]
We start by specifying the inverse of \( \mathcal{B} \),
\[ \mathcal{B}^{-1}(t) = \mathcal{E} e^{-\int dt' k \partial_k} \]
and stating
\[ \frac{d}{dt} \mathcal{B}^{-1}(t) = -q(E(t)) \partial_k \mathcal{E} e^{-\int dt' k \partial_k} \]

where we have used \( \hat{k}(t) = \partial_t (k-A(t)) = -\hat{A}(t) = q(E(t)) \) for a homogeneous electric field. We apply \( \mathcal{B}^{-1}(t) \) to the left of the \( g \) dynamics, Eq. (39), and obtain (suppressing the time dependence of \( \mathcal{B} \))
\[ \mathcal{B}^{-1} \frac{d}{dt} g_{nn'}(k; t) - \epsilon_{nn'}(k) \mathcal{B}^{-1} g_{nn'}(k; t) = E(t) \sum_{n} \left( \mathcal{B}^{-1} g_{n|k}(k) \right) d_{nn'}(k) - d_{nn'}(k) \mathcal{B}^{-1} g_{nn'}(k; t). \]
We insert Eq. (D3) and obtain
\[ \left( \frac{d}{dt} + i q E(t) \partial_k - \epsilon_{n'n'}(k) \right) \mathcal{B}^{-1} \varrho_{n'n'}(k; t) = \]
\[ E(t) \sum_{n} \left( \mathcal{B}^{-1} \varrho_{n}(k; t) \right) d_{n'n'}(k) - d_{n'n}(k) \mathcal{B}^{-1} \varrho_{n'n'}(k; t). \]  
(D4)

The EoM for $\mathcal{B}^{-1} \varrho_{n'n'}(k; t)$ in Eq. (D4) is identical to the EoM of $\rho_{n'n'}^{(B)}(k; t)$ in Eq. (47) and we conclude
\[ \mathcal{B}^{-1}(t) \varrho_{n'n'}(k; t) = \rho_{n'n'}^{(B)}(k; t). \]  
(D5)

Eq. (49) follows.

**Appendix E: Dynamical polarization $P$ in $k$-dependent basis**

In the main text, we derive the dynamical polarization in a Bloch basis with $k$-independent lattice-periodic part, see Eq. (53). In this appendix, we compute the polarization as expectation value of the dipole operator $\mathbb{q} \mathbb{r}$ in the stationary Bloch basis $|n k\rangle$ from (15) as [20]
\[ P(t) = \frac{1}{V} \sum_{\alpha, \beta} \langle \alpha | \mathbb{q} \mathbb{r} | \beta \rangle \rho_{\alpha \beta}(t) \]
\[ = \frac{1}{V} \sum_{n n'} \sum_{k' k} \langle n k | \mathbb{q} | n' k' \rangle \rho_{n'n'}(k' k; t), \]  
(E1)

with $V$ denoting the normalization volume. We keep the full $k$-dependence of
\[ \rho_{n'n'}(k' k; t) = \langle n k | \rho(t) | n' k' \rangle \]
in (E1) to properly account for $k$-derivatives of dipole matrix elements later on.

We evaluate the dipole matrix element $\langle n k | \mathbb{q} | n' k' \rangle$ appearing in the polarization (52) adopting (B1) as
\[ \langle n k | \mathbb{q} | n' k' \rangle = \frac{2\pi}{V} \langle n k | i \partial_k e^{-i(k' - k)\mathbb{r}} \delta(k' - k) | n' k' \rangle. \]  
(E2)

With (54) and results from Appendix A, we obtain
\[ P(t) = \frac{q}{V} \sum_{n n'} \int_{\text{BZ}} \frac{dk'}{(2\pi)^d} \rho_{n'n'}(k' k; t) \]
\[ \times \langle n k | i \partial_k e^{-i(k' - k)\mathbb{r}} \delta(k' - k) | n' k' \rangle. \]
\[ = \frac{q}{V} \sum_{n n'} \int_{\text{BZ}} \frac{dk'}{(2\pi)^d} \left( \langle n k | \partial_k | n' k' \rangle \rho_{n'n}(k k; t) \right) \]
\[ + \langle n k | n' k \rangle \frac{\partial \rho_{n'n}(k' k; t)}{\partial k'} |_{k' \rightarrow k}. \]  
(E3)

where integration by parts has been used to arrive at the last equation. We define
\[ \rho_{n'n}(k; t) := \langle c_{n'k} \partial_k c_{n'k} \rangle = \frac{\partial \rho_{n'n}(k' k; t)}{\partial k'} |_{k' \rightarrow k}. \]  
(E4)

so that
\[ P = \sum_{n n'} \int_{\text{BZ}} \frac{dk}{(2\pi)^d} \left( d_{n'n'}(k) \cdot \rho_{n'n}(k; t) + i q \delta_{n'n} \rho_{n'n}(k; t) \right) \]  
(E5)

recalling
\[ d_{n'n'}(k) = \langle n k | i q \partial_k | n' k \rangle \]  
(E6)

and abbreviated
\[ \rho_{n'n'}(k; t) := \rho_{n'n}(k k; t) = \langle n k | \rho(t) | n' k \rangle \]
\[ = \langle n k | \rho(t) | n' k \rangle. \]  
(A9)

The time derivative $\partial_t P$ of the polarization is needed for evaluating the emission (51) and is given by
\[ \partial_t P = \sum_{n n'} \int_{\text{BZ}} \frac{dk}{(2\pi)^d} \left( d_{n'n'}(k) \cdot \rho_{n'n}(k; t) + i q \delta_{n'n} \rho_{n'n}(k; t) \right) \]
\[ = \int_{\text{BZ}} \frac{dk}{(2\pi)^d} \text{Tr}_n \left( d(k) \rho(k; t) + i q \partial_t \rho(k; t) \right) \]  
(E8)

After inserting the EoM (40), $i \partial_t \rho(k; t) = [h(k; t), \rho(k; t)]$ in the Coulomb gauge in the right-hand side of (E4) the second term of (E8) contributes with
\[ i \sum_{n} \rho_{nn}(k; t) = \sum_{n n'} (\partial_k h_{nn'}(k, t)) \rho_{n'n}(k; t) \]  
(E9)

so that the coupling to $\rho(k; t)$ drops out due to cyclic invariance of the trace. Recalling the EoM for $\rho(t)$ we arrive at:
\[ \partial_t P = \sum_{n n'} \int_{\text{BZ}} \frac{dk}{(2\pi)^d} \left( [-i d(k), h(k; t)]_{nn'} \right) \]
\[ + q \partial_k h_{nn'}(k; t) \rho_{n'n}(k; t) \]
\[ = q \sum_{n n'} \int_{\text{BZ}} \frac{dk}{(2\pi)^d} \langle n k | \frac{\partial h(k; t)}{\partial k} | n' k \rangle \rho_{n'n}(k; t) \]
\[ = q \int_{\text{BZ}} \frac{dk}{(2\pi)^d} \text{Tr}_n \left[ \frac{\partial h(k; t)}{\partial k} \rho(k; t) \right] \]  
(E10)

The last line uses
\[ \partial_k h_{nn'}(k; t) = \partial_k \langle n k | h(k; t) | n' k \rangle \]
\[ = \langle n k | \partial_k h | n' k \rangle + \langle n k | \partial_k h | n' k \rangle + \langle n k | \partial_k h | n' k \rangle. \]  
(E11)

With Eq. (E10), we arrive at the same result as in Eq. (58) that has obtained using the Bloch basis with $k$-independent lattice-periodic part.
Appendix F: Proof of Eq. (65)

For the proof of Eq. (65),
\[ \langle n\k | \frac{\partial h^{\text{in}}(\k)}{\partial \k} | n\k' \rangle = \delta_{n'n'} \frac{\partial \epsilon_n(\k)}{\partial \k} + \frac{i}{q} \epsilon_{n'n'}(\k) d_{n'n'}(\k), \]
we execute
\[ \delta_{n'n'} \partial_\k \epsilon_n(\k) = \partial_\k \langle n\k | h^{\text{in}}(\k) | n\k' \rangle \]
\[ = \langle n\k | h^{\text{in}} \partial_\k | n\k' \rangle + \langle n\k | \frac{\partial h^{\text{in}}}{\partial \k} | n\k' \rangle + \langle n\k | \partial_\k h^{\text{in}} | n\k' \rangle \]
\[ = \epsilon_n(\k) \langle n\k | \partial_\k | n\k' \rangle + \langle n\k | \frac{\partial h^{\text{in}}}{\partial \k} | n\k' \rangle - \epsilon_{n'}(\k) \langle n\k | \partial_\k | n\k' \rangle \]
\[ = -\epsilon_n(\k) \frac{i}{q} d_{n'n'}(\k) + \langle n\k | \frac{\partial h^{\text{in}}}{\partial \k} | n\k' \rangle + \epsilon_{n'}(\k) \frac{i}{q} d_{n'n'}(\k), \]
where we have used \( \langle n\k | \partial_\k^{\dagger} | n'\k \rangle = -\langle n\k | \partial_\k | n'\k \rangle \).
Eq. (65) follows with \( \epsilon_{n'n'}(\k) = \epsilon_{n'}(\k) - \epsilon_n(\k). \)
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The Coulomb gauge is defined as $\nabla \cdot \mathbf{A} = 0$. In our application, source terms for generating electric fields are absent, i.e. $\Delta \Phi = 0$ and $\delta \mathbf{E} = 0$. In principle, a gauge-degree of freedom is left in this case. It implies possibilities for alternative representations, e.g., $\Phi(t) = -r\mathbf{E}(t)$ with the longitudinal component of $\mathbf{A}$ being independent of time ('length gauge') or $\mathbf{A}(t) = -q \int_{-\infty}^{t} \mathrm{d}t' \mathbf{E}(t')$ with $\Phi$ being independent of position ('velocity gauge'); [87] evaluating the expression $\mathbf{E} = -\nabla \Phi - \mathbf{A}/q$ in either gauge, the same electric field is reproduced. The representation of electric potential via the length gauge frequently occurs in the context of dipole expansions. In the literature, the velocity gauge and the Coulomb gauge are often identified with each other; for further discussion see Ref. 89.

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Note that due to translational invariance, in (23) only diagonal matrix elements with $\mathbf{k} = \mathbf{k}'$ appear and an additional sum $\sum_{\mathbf{k}'}$ is absent. We illustrate in Appendix B, (B3)-(B4) that contributions from off-diagonals $\mathbf{k} \neq \mathbf{k}'$ vanish.
We have discussed the Coulomb gauge (that is also referred to as velocity gauge) and dipole gauge (that is also referred to as length gauge) in footnote 81. The eigenstates of a Hamiltonian in both gauges are connected by a space-time dependent transformation \cite{104, 105} as it is also used in recent work \cite{87} focusing on dynamics of Bloch electrons. The space-dependence of this transformation turns into a derivative in $k$ such that eigenstates of a Hamiltonian in both gauges transform via the Boost operator $\mathcal{B}(t)$. As consequence, the density matrix in both gauges transforms as in Eq. (45).

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