Bloch-electron dynamics under the influence of a quantized radiation field

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A theory is described for a Bloch electron accelerating in a homogeneous external electric field of arbitrary time dependence while interacting with a quantized radiation field. The external electric field is described in the vector potential gauge. The quantum radiation field is described by the free space quantized electromagnetic field in the Coulomb gauge. The instantaneous eigenstates for the Bloch Hamiltonian are introduced as basis states to analyze the Bloch dynamics to all orders in the external electric field; as well, the eigenstates of the free quantum electromagnetic field are utilized as a component of the full basis set to develop a direct time-dependent solution of the Schrödinger equation. As an alternative consideration, the Glauber displacement operator is utilized to transform the original problem to a canonical form. For both the initial and transformed scenarios considered, the first-order solution to the Schrödinger equation is obtained and used to calculate the Bloch electric current in a form useful for studying the spectral content of solids. It is found that the Glauber transformed Hamiltonian and subsequent quantum dynamics is quite effective in providing solid-state band information in general and as noted in the first-order calculated Bloch-electron current.

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I. INTRODUCTION

Bloch-electron dynamics in external electric fields has been a subject of intense interest from the early development of solid-state physics [1]. Moreover, the modern development of band-engineered superlattices (SLs), tailored periodic structures, and low-dimensional materials (LDMs) has further stimulated a host of electric-field-mediated transport and optical absorption phenomena from SLs and quantum well (QW) nanostructures where the low-dimensional band gaps and bandwidths are typically several orders of magnitude smaller than those of bulk solids [2]. The band parameters of SLs, QWs, and LDMs give rise to transport with extreme, even ballistic, mean-free paths and radiation properties in the infrared and submillimeter wave range [3].

Most recently, there has been intense activity generated with the advent of picosecond to attosecond time-resolved technologies [4]. This has led to exciting new probe dynamics relevant to atomic and solid-state materials, and has stimulated new phenomena including high harmonic generation (HHG) [5]. With regard to the solid state, time-resolved methods draw attention to methodologies for addressing Bloch-electron dynamics in time-dependent radiation fields. In the typical theoretical approach for solids, the exciting radiation field is treated as a classical, time-dependent radiation field [6]. Yet, in experimental situations involving such phenomena as HHG, especially in the high-field intensity regime, the presence of high photon density dictates treating the *quantum nature* [6–8] of the exciting field as observable [9]. Thus, in this work, attention is focused on Bloch-electron dynamics in a quantized radiation field rather than a classical radiation field. The Bloch-electron current is calculated in considering the manifestation of useful spectral properties.

In Sec. II, the Hamiltonian for an accelerated Bloch electron in the free-space quantum electrodynamic field of interest is developed. The classical, time-dependent, homogeneous electric field is described in the vector potential gauge, and the free-space quantized electromagnetic radiation field is treated using the Coulomb gauge. In Sec. III, the single quantum field mode interaction with a Bloch electron is treated; the instantaneous eigenstates of the Bloch Hamiltonian are developed [10] as basis states to analyze the Bloch dynamics to all orders in the accelerating homogeneous electric field. Also, the eigenstates of the free quantum radiation field are utilized as a component of the complete basis set in developing the time-dependent solution to the Schrödinger equation. In addition, as an alternative formulation, the Glauber displacement [11] operator is utilized to transform the Schrödinger equation and Hamiltonian to an alternative form. In Sec. IV, it is shown that the current associated with the Glauber transformed case is solely dependent upon the Bloch band parameters. A first-order solution to the Schrödinger equation is obtained and utilized in the calculation of the Bloch-electron current in a form useful for studying the spectral content of solids. Section V provides a summary of results and conclusions. Seven Appendixes are included to supplement detail mathematical discussions as noted.

II. BLOCH HAMILTONIAN IN MULTIMODE ELECTROMAGNETIC FIELD

The Hamiltonian for a single electron in a periodic crystal potential $V_c(\mathbf{r})$ subject to an external time-varying

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electromagnetic field is

$$\hat{H}(\mathbf{r},\hat{\mathbf{p}},t)=\frac{1}{2m_e}\left[\hat{\mathbf{p}}-\frac{e}{c}\hat{\mathbf{A}}(\mathbf{r},t)\right]^2+V_c(\mathbf{r})+\hat{H}_r. \quad (1)$$

Here, e is the electron charge (that is, $e=-|e|$) and $\hat{\mathbf{p}}=-i\hbar\nabla_{\mathbf{r}}$ is the electron momentum operator, m_e is the free-electron mass, $\mathbf{r}$ is the space coordinate, $\hat{\mathbf{A}}(\mathbf{r},t)=\mathbf{A}_c(t)+\hat{\mathbf{A}}_r(\mathbf{r})$ is the total vector potential for the external homogeneous electric field, $\mathbf{E}(t)$, and for the free quantized radiation field described by the Hamiltonian $\hat{H}_r$. The vector potential $\mathbf{A}_c(t)$ is given by

$$\mathbf{A}_c(t)=-c\int_{t_0}^{t}\mathbf{E}(t')dt', \quad (2a)$$

where c is the speed of light in a vacuum, and t_0 is the time when the external electric field is turned on. The vector potential $\hat{\mathbf{A}}_r(\mathbf{r})$ for the free multimode quantized radiation field is given as

$$\hat{\mathbf{A}}_r(\mathbf{r})=\left(\frac{2\pi\hbar c}{V}\right)^{1/2}\sum_{\mathbf{q},\lambda}\frac{1}{q^{1/2}}(\hat{a}_{\mathbf{q}\lambda}\boldsymbol{\lambda}e^{i\mathbf{q}\cdot\mathbf{r}}+\hat{a}^{\dagger}_{\mathbf{q}\lambda}\boldsymbol{\lambda}^*e^{-i\mathbf{q}\cdot\mathbf{r}}). \quad (2b)$$

Here, $\boldsymbol{\lambda}$ is the two-component polarization [12] vector for each radiation mode of frequency $\omega_q=cq$ $(q=|\mathbf{q}|)$, the free-space photon dispersion with wave vector $\mathbf{q}$, V is the volume of the system, and $\hbar$ is the Planck constant divided with 2π. $\hat{\mathbf{A}}_r$ satisfies the Coulomb gauge so that $\nabla_{\mathbf{r}}\cdot\hat{\mathbf{A}}_r=0$ or $\boldsymbol{\lambda}\cdot\mathbf{q}=0$ for each polarization direction. Finally, $\hat{H}_r$ for the quantized radiation field is given by

$$\hat{H}_r=\sum_{\mathbf{q},\lambda}\hbar\omega_q\left(\hat{a}^{\dagger}_{\mathbf{q}\lambda}\hat{a}_{\mathbf{q}\lambda}+\frac{1}{2}\right), \quad (3)$$

where in both $\hat{\mathbf{A}}_r$ and $\hat{H}_r$, $\hat{a}^{\dagger}_{\mathbf{q}\lambda}$ and $\hat{a}_{\mathbf{q}\lambda}$ are the well-known creation and annihilation boson operators for the quantum radiation field.

In substituting the vector potential $\hat{\mathbf{A}}(\mathbf{r},t)$ into Eq. (1), the Hamiltonian can be reexpressed as

$$\hat{H}=\hat{H}_0+\hat{H}_r+\hat{H}'_i, \quad (4)$$

where

$$\hat{H}_0=\frac{1}{2m_e}(\hat{\mathbf{p}}+\mathbf{p}_c)^2+V_c(\mathbf{r}), \quad (5)$$

$\hat{H}_r$ is given by Eq. (3), and $\hat{H}'_i$ is given as

$$\hat{H}'_i=\hat{H}_i+\hat{H}_A, \quad (6a)$$

where

$$\hat{H}_i=-\frac{e}{m_ec}\hat{\mathbf{A}}_r\cdot(\hat{\mathbf{p}}+\mathbf{p}_c), \quad (6b)$$

and

$$\hat{H}_A=\frac{e^2}{2m_ec^2}\hat{\mathbf{A}}_r^2. \quad (6c)$$

In solving the time-dependent Schrödinger equation associated with the Hamiltonian of Eq. (4),

$$i\hbar\frac{\partial}{\partial t}|\Psi(t)\rangle=\hat{H}|\Psi(t)\rangle, \quad (7)$$

we note that, in a natural approach, we use the instantaneous eigenstates of $\hat{H}_0$ and $\hat{H}_r$ as basis functions and look for a general solution of Eq. (7) in terms of the complete set of functions. For $\hat{H}_0$ of Eq. (5), we chose the *instantaneous eigenstates* of $\hat{H}_0$, the well-established states [10,13],

$$\hat{H}_0\psi_{n\mathbf{K}}(\mathbf{r},t)=\varepsilon_n[\mathbf{k}(t)]\psi_{n\mathbf{K}}(\mathbf{r},t), \quad (8a)$$

where $\mathbf{k}(t)=\mathbf{K}+\mathbf{k}_c(t), \mathbf{k}_c(t)=\mathbf{p}_c(t)/\hbar=(e/\hbar)\int_{t_0}^{t}\mathbf{E}(t')dt'$, $\varepsilon_n(\mathbf{K})$ represents the energy dispersion of the band n, and

$$\psi_{n\mathbf{K}}(\mathbf{r},t)=\frac{e^{i\mathbf{K}\cdot\mathbf{r}}}{V^{1/2}}u_{n\mathbf{k}(t)}(\mathbf{r},t), \quad (8b)$$

where V is the volume of the crystal. For $\hat{H}_r$ of Eq. (3), we have the well-known oscillator states $|\{n_{\mathbf{q}\lambda}\}\rangle$ given by

$$\hat{H}_r|\{n_{\mathbf{q},\lambda}\}\rangle=\sum_{\mathbf{q},\lambda}\hbar\omega_q|\{n_{\mathbf{q}\lambda}\}\rangle. \quad (8c)$$

Then, we formally look for $|\Psi(t)\rangle$ of Eq. (7) in the form

$$|\Psi(t)\rangle=\sum_{n,\mathbf{k}}\sum_{\{n_{\mathbf{q}\lambda}\}}A_{n\mathbf{k},\{n_{\mathbf{q}\lambda}\}}(t)\psi_{n\mathbf{K}}|\{n_{\mathbf{q}\lambda}\}\rangle \times e^{-\frac{i}{\hbar}\int_{t_0}^{t}dt'\{\varepsilon_n[\mathbf{k}(t')]+\sum_{\mathbf{q},\lambda}\hbar\omega_q n_{\mathbf{q}\lambda}\}}, \quad (9)$$

where the coefficients, $A_{n\mathbf{k},\{n_{\mathbf{q}\lambda}\}}(t)$, are found by substituting Eq. (9) into Eq. (7) and appropriately solving for $A_{n\mathbf{k},\{n_{\mathbf{q}\lambda}\}}(t)$. In this general formalism, we emphasize that Eq. (9) includes the sum over $\{n_{\mathbf{q}\lambda}\}$; this indicates that the general solution requires a sum *over all* the radiation modes of the quantum field. This ensures that all possible multiphoton processes are included in the general solution.

The Hamiltonian of Eq. (4) is in a convenient form for developing perturbation theory solutions to the Schrödinger equation since it orders the quantized field in powers of $\hat{\mathbf{A}}_r$. But this Hamiltonian is also quite sensitive to the type of polarization characterizing the radiation field. This was clearly demonstrated [14] for a single-mode analysis where the electron Hamiltonian was assumed to be that of a *free particle*; this referenced work makes use of the Glauber displacement operator to simplify the Hamiltonian associated with the free-particle case consideration. In our work, we make use of a similar approach with a single-mode analysis, but with the electron Hamiltonian defined for a single Bloch electron in a homogeneous electric field of arbitrary time dependence.

III. BLOCH DYNAMICS IN A SINGLE-MODE RADIATION FIELD

The Hamiltonian taken from Eq. (4) can be written in a form convenient for single-mode consideration as

$$\hat{H}(\mathbf{r},\hat{\mathbf{p}},t)=\frac{1}{2m_e}(\hat{\mathbf{p}}+\mathbf{p}_c)^2+V_c(\mathbf{r})+\hat{H}_r -\frac{e}{m_ec}\hat{\mathbf{A}}_r\cdot(\hat{\mathbf{p}}+\mathbf{p}_c)+\frac{e^2}{2m_ec^2}\hat{\mathbf{A}}_r^2. \quad (10)$$

Letting $\omega_q=\omega$ and $\hat{a}_{\mathbf{q}\lambda}=\hat{a}$, then $\hat{H}_r$ and $\hat{\mathbf{A}}_r$ reduce to

$$\hat{H}_r=\hbar\omega\left(\hat{a}^{\dagger}\hat{a}+\tfrac{1}{2}\right), \quad (11a)$$

and

$$\hat{\mathbf{A}}_r = \left(\frac{2\pi\hbar c}{Vq}\right)^{1/2}(\hat{a}\boldsymbol{\lambda}e^{i\mathbf{q}\cdot\mathbf{r}} + \hat{a}^\dagger\boldsymbol{\lambda}^* e^{-i\mathbf{q}\cdot\mathbf{r}}), \tag{11b}$$

respectively; in the Coulomb gauge, $\hat{\mathbf{p}}\cdot\hat{\mathbf{A}}_r = 0$. In Eq. (11b), $\boldsymbol{\lambda}$ is the polarization [12] vector for the radiation field with $|\boldsymbol{\lambda}|^2 = 1$. For linear polarization, in the $\hat{\mathbf{x}}$ direction, $\boldsymbol{\lambda}(\hat{\mathbf{x}}) = (1, 0)$; in the $\hat{\mathbf{y}}$ direction, $\boldsymbol{\lambda}(\hat{\mathbf{y}}) = (0, 1)$. For circular polarization, $\boldsymbol{\lambda}\cdot\boldsymbol{\lambda} = \boldsymbol{\lambda}^*\cdot\boldsymbol{\lambda}^* = 0$ and $|\boldsymbol{\lambda}|^2 = 1$, for right helicity, $\boldsymbol{\lambda}_R = (1, i)/\sqrt{2}$, and left helicity, $\boldsymbol{\lambda}_L = (1, -i)/\sqrt{2}$. General polarization algebra can also be established, but we focus here especially on *right circular polarization* for the remainder of this analysis.

For $\hat{\mathbf{A}}_r^2$, it follows from Eq. (11b) that

$$\hat{\mathbf{A}}_r^2 = \frac{4\pi\hbar c}{Vq}\left(\hat{a}^\dagger\hat{a} + \frac{1}{2}\right). \tag{12}$$

We note that the radiation field Hamiltonian $\hat{H}_r$, given in Eq. (11a), and $\hat{\mathbf{A}}_r^2$ terms of Eqs. (6c) and (12) combine to give

$$\hat{\mathcal{H}}_r \equiv \hat{H}_r + \hat{H}_A = \hbar\Omega\left(\hat{a}^\dagger\hat{a} + \tfrac{1}{2}\right), \tag{13}$$

where

$$\Omega = \omega\left(1 + \frac{\omega_p^2}{2\omega^2}\right), \tag{14}$$

and $\omega_p = (4\pi e^2/m_e V)^{1/2}$ is the plasma frequency [15]. Thus, the single-mode Hamiltonian which follows from Eq. (10) for circular polarization is

$$\hat{H}(\mathbf{r}, \hat{\mathbf{p}}, t) = \hat{H}_0 + \hbar\Omega\left(\hat{a}^\dagger\hat{a} + \tfrac{1}{2}\right) + \hat{H}', \tag{15a}$$

$$\hat{H}' = -D_0(\hat{\mathbf{p}} + \mathbf{p}_c)\cdot(\hat{a}\boldsymbol{\lambda}e^{i\mathbf{q}\cdot\mathbf{r}} + \hat{a}^\dagger\boldsymbol{\lambda}^* e^{-i\mathbf{q}\cdot\mathbf{r}}), \tag{15b}$$

where $D_0 = (e/m_e)(2\pi\hbar/V\omega)^{1/2}$, and $\boldsymbol{\lambda}$ belongs to $\boldsymbol{\lambda}_R$ and $\boldsymbol{\lambda}_L$. To solve the time-dependent Schrödinger equation, Eq. (7), we use two different methods which are described below.

A. Direct solution of time-dependent Schrödinger equation for the Hamiltonian $\hat{H}$

For our purpose here, it is convenient to reexpress the Hamiltonian $\hat{H}$ in Eq. (15a) in the form

$$\hat{H}(\mathbf{r}, \hat{\mathbf{p}}, t) = \hat{H}_0 + \hat{H}', \tag{16a}$$

where we have redesignated $\hat{H}_0$ of Eq. (5) as

$$\hat{H}_0 = \frac{1}{2m_e}(\hat{\mathbf{p}} + \mathbf{p}_c)^2 + V_c(\mathbf{r}) + \hbar\Omega\left(\hat{a}^\dagger\hat{a} + \frac{1}{2}\right), \tag{16b}$$

and $\hat{H}'$ is given in Eq. (15b). We look for the solution to the time-dependent Schrödinger equation, Eq. (7), using the instantaneous eigenstates of $\hat{H}_0$ in Eq. (16b) as basis states. These states are described by the band wave functions, $\psi_{n\mathbf{K}}(\mathbf{r}, t)$ with eigenvalues $\varepsilon_n[\mathbf{k}(t)]$, given in Eqs. (8a) and (8b); also, the eigenvectors $|m\rangle$ and eigenvalues $E_m = \hbar\Omega(m + \frac{1}{2})$ of the harmonic-oscillator Hamiltonian in Eq. (13). Then, we can express the solution to Eq. (7) as

$$|\Psi(t)\rangle = \sum_{n,\mathbf{k}}\sum_m A_{n\mathbf{k}}(m, t)\psi_{n\mathbf{K}}(\mathbf{r}, t)|m\rangle e^{-\frac{i}{\hbar}\int_{t_0}^t\{\varepsilon_n[\mathbf{k}(\tau)]+E_m\}d\tau}. \tag{17}$$

Putting $|\Psi(t)\rangle$ into Eq. (7) and taking the scalar products with $\psi_{n\mathbf{K}}(\mathbf{r}, t)|m\rangle$, using the orthogonality properties of both $\psi_{n\mathbf{K}}$ and $|m\rangle$, we obtain

$$\begin{aligned}\dot{A}_{n\mathbf{k}}(m, t) = &-\frac{1}{i\hbar}\sum_{n'\neq n} A_{n'\mathbf{k}}(m, t)\mathbf{F}(t)\cdot\mathbf{R}_{nn'}(\mathbf{k})e^{\frac{i}{\hbar}\int_{t_0}^t[\varepsilon_n(\mathbf{k})-\varepsilon_{n'}(\mathbf{k})]d\tau}\\ &+\frac{1}{i\hbar}\sum_{n',\mathbf{k}'}\sum_{m'} A_{n'\mathbf{k}'}(m', t)\hat{H}'_{n\mathbf{k},n'\mathbf{k}'}(m, m', t)e^{\frac{i}{\hbar}\int_{t_0}^t[\varepsilon_n(\mathbf{k})-\varepsilon_{n'}(\mathbf{k}')+E_m-E_{m'}]d\tau},\end{aligned} \tag{18a}$$

where

$$\mathbf{F}(t) = e\mathbf{E}(t), \quad \mathbf{R}_{nn'}(\mathbf{k}) = \frac{i}{\Omega_c}\int_{\Omega_c} u_{n\mathbf{k}}^*(\mathbf{r})\nabla_{\mathbf{k}}u_{n'\mathbf{k}}(\mathbf{r})d\mathbf{r}, \tag{18b}$$

and Ω_c is the volume of the unit cell. For matrix elements of $\hat{H}'$ given in Eq. (15b), we find using the chosen basis set of functions that

$$\hat{H}'_{n\mathbf{k},n'\mathbf{k}'}(m, m', t) = -D_0[\hat{a}_{mm'}\boldsymbol{\lambda}\cdot(\hat{\mathbf{p}} + \mathbf{p}_c)_{n\mathbf{k},n'\mathbf{k}'}\delta_{\mathbf{k},\mathbf{k}'+\mathbf{q}} + \hat{a}^\dagger_{mm'}\boldsymbol{\lambda}^*\cdot(\hat{\mathbf{p}} + \mathbf{p}_c)_{n\mathbf{k},n'\mathbf{k}'}\delta_{\mathbf{k},\mathbf{k}'-\mathbf{q}}], \tag{19}$$

where $\hat{a}_{mm'} = (m')^{1/2}\delta_{m,m'-1}$, $\hat{a}^\dagger_{mm'} = (m'+1)^{1/2}\delta_{m,m'+1}$, and the terms $\delta_{\mathbf{k},\mathbf{k}'\pm\mathbf{q}}$ come from the radial contribution from the matrix elements of Eq. (19). From Eq. (18a), it follows after integration that

$$\begin{aligned}A_{n\mathbf{k}}(m, t) = &-\frac{1}{i\hbar}\sum_{n'\neq n}\int_{t_0}^t dt' A_{n'\mathbf{k}}(m, t')\mathbf{F}(t')\cdot\mathbf{R}_{nn'}(\mathbf{k})e^{\frac{i}{\hbar}\int_{t_0}^{t'}[\varepsilon_n(\mathbf{k})-\varepsilon_{n'}(\mathbf{k})]dt'}\\ &+\frac{1}{i\hbar}\sum_{n',\mathbf{k}'}\sum_{m'}\int_{t_0}^t dt' A_{n'\mathbf{k}'}(m', t')\hat{H}'_{n\mathbf{k},n'\mathbf{k}'}(m, m', t')e^{\frac{i}{\hbar}\int_{t_0}^{t'}[\varepsilon_n(\mathbf{k})-\varepsilon_{n'}(\mathbf{k}')+E_m-E_{m'}]d\tau}.\end{aligned} \tag{20}$$

Seeking an approximate solution to Eq. (20) in the form

$$A_{n\mathbf{k}}(m,t) \simeq \delta_{n,n_0}\delta_{\mathbf{K},\mathbf{K}_0}\delta_{m,m_0} + A^{(1)}_{n\mathbf{k}}(m,t), \tag{21a}$$

where $A_{n\mathbf{k}}(m,t_0) = \delta_{n,n_0}\delta_{\mathbf{K},\mathbf{K}_0}\delta_{m,m_0}$ and $A^{(1)}_{n\mathbf{k}}(m,t_0) = 0$ correspond to the initial state $\psi_{n_0\mathbf{K}_0}|m_0\rangle$ at $t = t_0$, we find after insertion into right-hand side of Eq. (20) and integration

$$\begin{aligned} A^{(1)}_{n\mathbf{k}}(m,t) \simeq & -\frac{1}{i\hbar}\int_{t_0}^{t} dt'\mathbf{F}(t')\cdot\mathbf{R}_{nn_0}(\mathbf{k})e^{\frac{i}{\hbar}\int_{t_0}^{t'}[\varepsilon_n(\mathbf{k}_0)-\varepsilon_{n_0}(\mathbf{k}_0)]d\tau}\delta_{\mathbf{k},\mathbf{k}_0}\delta_{m,m_0} \\ & - m_0^{1/2}\frac{D_0}{i\hbar}\int_{t_0}^{t} dt'\boldsymbol{\lambda}\cdot(\hat{\mathbf{p}}+\mathbf{p}_c)_{n\mathbf{k},n_0\mathbf{k}_0}e^{\frac{i}{\hbar}\int_{t_0}^{t'}[\varepsilon_n(\mathbf{k}_0+\mathbf{q})-\varepsilon_{n_0}(\mathbf{k}_0)-\hbar\Omega]d\tau}\delta_{\mathbf{k},\mathbf{k}_0+\mathbf{q}}\delta_{m,m_0-1} \\ & - (m_0+1)^{1/2}\frac{D_0}{i\hbar}\int_{t_0}^{t} dt'\boldsymbol{\lambda}^*\cdot(\hat{\mathbf{p}}+\mathbf{p}_c)_{n\mathbf{k},n_0\mathbf{k}_0}e^{\frac{i}{\hbar}\int_{t_0}^{t'}[\varepsilon_n(\mathbf{k}_0-\mathbf{q})-\varepsilon_{n_0}(\mathbf{k}_0)+\hbar\Omega]d\tau}\delta_{\mathbf{k},\mathbf{k}_0-\mathbf{q}}\delta_{m,m_0+1}. \end{aligned} \tag{21b}$$

Here, $\mathbf{k}_0(t) = \mathbf{K}_0 = \mathbf{p}_c(t)/\hbar$. In Eq. (21b), the first term is the well-known [10] Zener tunneling coefficient, and the second and third terms are the single-photon absorption and emission coefficient. We therefore express the result of Eq. (21b) as

$$A^{(1)}_{n\mathbf{k}}(m,t) = A^{(Z)}_{n\mathbf{k}}(t) + A^{(a)}_{n\mathbf{k}}(m,t) + A^{(e)}_{n\mathbf{k}}(m,t). \tag{22}$$

Thus, the solution to the Schrödinger equation (7) is found to first order by putting Eq. (22) into Eq. (17). We note that in the absorption and emission terms, when considering electron-photon dynamics in Bloch bands, the values of photon vector $\mathbf{q}$ are orders of magnitude smaller than Bloch $\mathbf{k}$ vector well into the UV. Thus, $|\mathbf{q}| \ll |\mathbf{k}|$ may be suppressed relative to $|\mathbf{k}|$ (see Appendix A).

B. Alternative approach based on unitary transformation

In this section, we consider an alternative approach to solving Eqs. (7) and (16a) by applying the Glauber-type [11] unitary transformation $\Psi = \hat{D}\Phi$, $\hat{\mathcal{H}} = \hat{D}^{-1}\hat{H}\hat{D}$ ($\hat{D}^\dagger = \hat{D}^{-1}$ and $\hat{D}^{-1}\hat{D} = \hat{D}\hat{D}^{-1} = \hat{I}$), where

$$\hat{D} = e^{\hat{\sigma}\hat{a}^\dagger - \hat{\sigma}^\dagger\hat{a}}, \quad \hat{D}^\dagger = e^{\hat{\sigma}^\dagger\hat{a} - \hat{\sigma}\hat{a}^\dagger}, \tag{23a}$$

but where $\hat{\sigma}, \hat{\sigma}^\dagger$ are arbitrary operators that are independent of $\hat{a}$, $\hat{a}^\dagger$ and therefore commute with $\hat{a}$ and $\hat{a}^\dagger$. With respect to $\hat{a}$ and $\hat{a}^\dagger$, $\hat{D}$ possesses the transformation properties [11]

$$\hat{D}^{-1}\hat{a}\hat{D} = \hat{a} + \hat{\sigma}, \quad \hat{D}^{-1}\hat{a}^\dagger\hat{D} = \hat{a}^\dagger + \hat{\sigma}^\dagger. \tag{23b}$$

Thus, in using $\hat{D}, \hat{D}^\dagger$ in Eq. (16a), we obtain

$$\hat{\mathcal{H}} \equiv \hat{D}^{-1}\hat{H}\hat{D} = \hat{D}^{-1}\hat{H}_0\hat{D} + \hat{D}^{-1}\hat{H}'\hat{D}. \tag{24}$$

In applying the transformation $\hat{D}$ with properties from Eq. (23b) to $\hat{H}_0$ and $\hat{H}'$ specified in Eqs. (16b) and (15b), respectively, and then choosing $\hat{\sigma}$ and $\hat{\sigma}^\dagger$ to eliminate the linear dependence of $\hat{a}$ and $\hat{a}^\dagger$ in the transformed $\hat{H}$, we find that Eq. (24) becomes

$$\hat{\mathcal{H}} = \hat{\mathcal{H}}_0 - \hbar\Omega\hat{\sigma}\hat{\sigma}^\dagger, \tag{25a}$$

where

$$\hat{\mathcal{H}}_0 = \hat{D}^{-1}\hat{H}_0\hat{D}, \tag{25b}$$

and the required choice for $\hat{\sigma}, \hat{\sigma}^\dagger$ is

$$\hat{\sigma} = \frac{D_0}{\hbar\Omega}(\hat{\mathbf{p}}+\mathbf{p}_c)\cdot\boldsymbol{\lambda}^* e^{-i\mathbf{q}\cdot\mathbf{r}}, \quad \hat{\sigma}^\dagger = \frac{D_0}{\hbar\Omega}(\hat{\mathbf{p}}+\mathbf{p}_c)\cdot\boldsymbol{\lambda}e^{i\mathbf{q}\cdot\mathbf{r}}. \tag{26}$$

In applying the transformation properties of $\hat{D}$ in Eqs. (23b) to (24), we note, as seen in Eq. (26), that $\hat{\sigma}$ and $\hat{\sigma}^\dagger$ will generally depend on noncommuting variables $\mathbf{r}$ and $\hat{\mathbf{p}}$. The quantum nature of this noncommutation must be taken into account when evaluating $\hat{\sigma}\hat{\sigma}^\dagger$ and $\hat{\mathcal{H}}_0$ in the unfolding quantum dynamical equations. The general commutation properties of $\hat{\sigma}, \hat{\sigma}^\dagger$ are discussed in Appendix B.

However, we note that when considering electron-photon dynamics in Bloch bands of crystals, the values of photon wave vector $\mathbf{q}$ of interest in this work are orders of magnitude smaller than the Bloch $\mathbf{K}$ vector. Thus, we suppress $\mathbf{q}$ relative to $\mathbf{k}$ (Appendix A), and we therefore ignore $e^{\pm i\mathbf{q}\cdot\mathbf{r}}$ in $\hat{H}'$ of Eq. (15b). This approximation is essentially equivalent [16] to the dipole approximation for atoms; it differs from the atomic case in that for solids the effective dipole approximation becomes transparent through momentum transition matrix elements over the Bloch states in question [see Eqs. (19), (21b), and (22), for example]. In this work, we invoke the dipole approximation by letting $\mathbf{q} \to 0$ (see Appendix A).

In general, we note that using $\hat{\sigma}, \hat{\sigma}^\dagger$ of Eq. (26), $\hat{D}$ can be reexpressed as

$$\hat{D} = e^{(\hat{\mathbf{p}}+\mathbf{p}_c)\cdot\hat{\boldsymbol{\alpha}}}, \tag{27a}$$

where

$$\hat{\boldsymbol{\alpha}} = \frac{D_0}{\hbar\Omega}(\boldsymbol{\lambda}^*\hat{a}^\dagger e^{-i\mathbf{q}\cdot\mathbf{r}} - \boldsymbol{\lambda}\hat{a}e^{i\mathbf{q}\cdot\mathbf{r}}). \tag{27b}$$

But, invoking the dipole approximation, we let $\mathbf{q} \to 0$, so that $\hat{\boldsymbol{\alpha}}$ of Eq. (27b) becomes $\hat{\boldsymbol{\alpha}}_0$, where

$$\hat{\boldsymbol{\alpha}}_0 = \frac{D_0}{\hbar\Omega}(\boldsymbol{\lambda}^*\hat{a}^\dagger - \boldsymbol{\lambda}\hat{a}), \tag{27c}$$

with $\hat{\boldsymbol{\alpha}}_0^\dagger = -\hat{\boldsymbol{\alpha}}_0$. Then $\hat{D}$ of Eq. (27a) becomes $\hat{\mathcal{D}}_0$, where

$$\hat{\mathcal{D}}_0 = e^{(\hat{\mathbf{p}}+\mathbf{p}_c)\cdot\hat{\boldsymbol{\alpha}}_0}, \quad \hat{\mathcal{D}}_0^\dagger = e^{-(\hat{\mathbf{p}}+\mathbf{p}_c)\cdot\hat{\boldsymbol{\alpha}}_0}. \tag{28}$$

We find from Eqs. (26) and (B7) that, when $\mathbf{q} \to 0$, $\hat{\sigma} \to \hat{\sigma}_0$, and $\hat{\sigma}^\dagger \to \hat{\sigma}_0^\dagger$, where

$$\hat{\sigma}_0 = \frac{D_0}{\hbar\Omega}\boldsymbol{\lambda}^*\cdot(\hat{\mathbf{p}}+\mathbf{p}_c), \quad \hat{\sigma}_0^\dagger = \frac{D_0}{\hbar\Omega}\boldsymbol{\lambda}\cdot(\hat{\mathbf{p}}+\mathbf{p}_c), \tag{29}$$

so that the Hamiltonian $\hat{\mathcal{H}}$ of Eqs. (24) and (25a) becomes

$$\begin{aligned} \hat{\mathcal{H}} = & \frac{1}{2m_e}(\hat{\mathbf{p}}+\mathbf{p}_c)^2 + V_c(\mathbf{r}+i\hbar\hat{\boldsymbol{\alpha}}_0) + \hbar\Omega\left(\hat{a}^\dagger\hat{a}+\frac{1}{2}\right) \\ & - \frac{D_0^2}{\hbar\Omega}(\hat{\mathbf{p}}+\mathbf{p}_c)\cdot\boldsymbol{\lambda}^*\,(\hat{\mathbf{p}}+\mathbf{p}_c)\cdot\boldsymbol{\lambda}. \end{aligned} \tag{30}$$

The Schrödinger equation (7) is transformed via $|\Psi(t)\rangle = \hat{\mathcal{D}}_0|\Phi(t)\rangle$ using $\hat{\mathcal{D}}_0$ of Eq. (28) into the equation

$$i\hbar\left[\frac{\partial}{\partial t} + \hat{\mathcal{D}}_0^\dagger \frac{\partial \hat{\mathcal{D}}_0}{\partial t}\right]|\Phi(t)\rangle = \hat{\mathcal{H}}|\Phi(t)\rangle. \tag{31}$$

From Eq. (28), with $\hat{\mathcal{D}}_0 = \hat{\mathcal{D}}_0(t)$, we find

$$\hat{\mathcal{D}}_0^\dagger \frac{\partial \hat{\mathcal{D}}_0}{\partial t} = \hat{\boldsymbol{\alpha}}_0 \cdot \dot{\mathbf{p}}_c \tag{32a}$$

and

$$\hat{\mathcal{D}}_0^\dagger V_c(\mathbf{r})\hat{\mathcal{D}}_0 = V_c(\mathbf{r} + i\hbar\hat{\boldsymbol{\alpha}}_0). \tag{32b}$$

In Eq. (32b), we have made use of the fact that $\hat{\mathcal{D}}_0$ from Eq. (28) commutes with $\hat{\mathbf{p}}$, and $[\mathbf{r}, \hat{\mathcal{D}}_0] = i\hbar\nabla_{\mathbf{p}}\hat{\mathcal{D}}_0 = i\hbar\hat{\boldsymbol{\alpha}}_0\hat{\mathcal{D}}_0$. Then, the Schrödinger equation transformation of Eq. (31) becomes

$$\begin{aligned} i\hbar\frac{\partial}{\partial t}&|\Phi(t)\rangle + i\hbar\hat{\boldsymbol{\alpha}}_0 \cdot \dot{\mathbf{p}}_c \\ &= \left\{\frac{1}{2m_e}(\hat{\mathbf{p}} + \mathbf{p}_c)^2 + V_c(\mathbf{r} + i\hbar\hat{\boldsymbol{\alpha}}_0) + \hbar\Omega\left(\hat{a}^\dagger\hat{a} + \frac{1}{2}\right)\right. \\ &\quad \left. - \frac{D_0^2}{\hbar\Omega}(\hat{\mathbf{p}} + \mathbf{p}_c)\cdot\boldsymbol{\lambda}^*\,(\hat{\mathbf{p}} + \mathbf{p}_c)\cdot\boldsymbol{\lambda}\right\}|\Phi(t)\rangle, \end{aligned} \tag{32c}$$

which can be written in the form similar to Eq. (7) as

$$i\hbar\frac{\partial}{\partial t}|\Phi(t)\rangle = (\hat{\mathcal{H}}_0 + \hat{\mathcal{H}}')|\Phi(t)\rangle, \tag{33a}$$

where

$$\hat{\mathcal{H}}_0 = \frac{1}{2m_e}(\hat{\mathbf{p}} + \mathbf{p}_c)^2 + V_c(\mathbf{r}) + \hbar\Omega\left(\hat{a}^\dagger\hat{a} + \frac{1}{2}\right) \tag{33b}$$

and

$$\begin{aligned} \hat{\mathcal{H}}' &= V_c(\mathbf{r} + i\hbar\hat{\boldsymbol{\alpha}}_0) - V_c(\mathbf{r}) \\ &\quad - \frac{D_0^2}{\hbar\Omega}(\hat{\mathbf{p}} + \mathbf{p}_c)\cdot\boldsymbol{\lambda}^*\,(\hat{\mathbf{p}} + \mathbf{p}_c)\cdot\boldsymbol{\lambda} - i\hbar\hat{\boldsymbol{\alpha}}_0 \cdot \dot{\mathbf{p}}_c. \end{aligned} \tag{33c}$$

As noted by comparison, Eqs. (16b) and (33b) are identical with respect to $\hat{H}_0$, so that we can adopt the same dual basis set; that is, instantaneous Bloch electron eigenstates and harmonic-oscillator eigenstates of Eq. (16b) in solving the transformed Schrödinger equation of Eq. (33a). However, Eq. (15b) of the original Hamiltonian and Eq. (33c) of the transformed Hamiltonian are quite different, with Eq. (33c) reflecting the transforming properties of the unitary operator, $\hat{\mathcal{D}}_0$; that is, the notable spatial shift of $\hat{\boldsymbol{\alpha}}_0$ in the crystal potential energy, $V_c(\mathbf{r} + i\hbar\hat{\boldsymbol{\alpha}}_0) - V_c(\mathbf{r})$, a shift which is reminiscent of the Henneberger-Kramers shift [17]. As well, this shift serves as a spatial optical inner probe of $V_c(\mathbf{r})$ and, together with the term $i\hbar\hat{\boldsymbol{\alpha}}_0 \cdot \dot{\mathbf{p}}_c$, represents an explicit measure of the competition between the Bloch and classical fields, mediated by the radiation field interaction through $\hat{\boldsymbol{\alpha}}_0$. Lastly, the $(\hat{\mathbf{p}} + \mathbf{p}_c)\cdot\boldsymbol{\lambda}^*\,(\hat{\mathbf{p}} + \mathbf{p}_c)\cdot\boldsymbol{\lambda}$ term, when expanded for circularly polarized $\boldsymbol{\lambda}, \boldsymbol{\lambda}^*$, results in $-\frac{D_0^2}{2\hbar\Omega}(\hat{\mathbf{p}} + \mathbf{p}_c)_\perp^2$, representing a contracted perpendicular component of the kinetic energy due to the quantum circular polarization.

We also note that in expressing $\hat{\mathcal{D}}_0$ of Eq. (28) in terms of $\hat{\sigma}_0$ of Eq. (29), it is easy to show using Eq. (23b) that $|\hat{\sigma}_0\rangle = \hat{\mathcal{D}}_0|0\rangle$ is a *coherent-like* state in that $\hat{a}|\hat{\sigma}_0\rangle = \hat{\sigma}_0|\hat{\sigma}_0\rangle$, where $\hat{\sigma}_0$ commutes with $\hat{a}$.

As noted, the interesting feature of $V_c(\mathbf{r} + i\hbar\hat{\boldsymbol{\alpha}}_0)$ arises when we observe the matrix elements with the chosen basis functions. For this purpose, representing $V_c(\mathbf{r})$ by its crystal Fourier representation

$$V_c(\mathbf{r}) = \sum_{\mathbf{G}} U_{\mathbf{G}} e^{i\mathbf{G}\cdot\mathbf{r}}, \quad U_{\mathbf{G}} = \frac{1}{\Omega_c}\int_{\Omega_c} V_c(\mathbf{r})e^{-i\mathbf{G}\cdot\mathbf{r}}d\mathbf{r}, \tag{34}$$

with $\mathbf{G}$ belonging to the reciprocal lattice, we see that

$$\begin{aligned} \langle m'|\psi_{n'\mathbf{K}'}|&V_c(\mathbf{r} + i\hbar\hat{\boldsymbol{\alpha}}_0)|\psi_{n\mathbf{K}}|m\rangle \\ &= \sum_{\mathbf{G}} U_{\mathbf{G}}\langle\psi_{n'\mathbf{K}'}|e^{i\mathbf{G}\cdot\mathbf{r}}|\psi_{n\mathbf{K}}\rangle\langle m'|e^{-\hbar\mathbf{G}\cdot\hat{\boldsymbol{\alpha}}_0)}|m\rangle. \end{aligned}$$

Since

$$\begin{aligned} \langle\psi_{n'\mathbf{K}'}|e^{i\mathbf{G}\cdot\mathbf{r}}|\psi_{n\mathbf{K}}\rangle &= \frac{1}{V}\int_{\mathrm{V}} u_{n'\mathbf{k}'}^* u_{n\mathbf{k}} e^{i(\mathbf{k}-\mathbf{k}'+\mathbf{G})\cdot\mathbf{r}}d\mathbf{r} \\ &= \frac{1}{\Omega_c}\int_{\Omega_c} u_{n'\mathbf{k}'}^* u_{n\mathbf{k}} d\mathbf{r}\,\delta_{\mathbf{k}'-\mathbf{k},\mathbf{G}}, \end{aligned}$$

so that $\mathbf{k}' - \mathbf{k} = \mathbf{G}$, noted as the electron-diffraction condition, we can write

$$\begin{aligned} \langle m'|\psi_{n'\mathbf{K}'}|&V_c(\mathbf{r} + i\hbar\hat{\boldsymbol{\alpha}}_0)|\psi_{n\mathbf{K}}|m\rangle \\ &= U_{\mathbf{k}'-\mathbf{k}} O_{n'\mathbf{k}',n\mathbf{k}}\langle m'|e^{-\hbar(\mathbf{k}'-\mathbf{k})\cdot\hat{\boldsymbol{\alpha}}_0}|m\rangle, \end{aligned} \tag{35a}$$

where

$$\begin{aligned} O_{n'\mathbf{k}',n\mathbf{k}} &= \frac{1}{\Omega_c}\int_{\Omega_c} u_{n'\mathbf{k}'}^*(\mathbf{r})u_{n\mathbf{k}}(\mathbf{r})d\mathbf{r}\,\delta_{\mathbf{k}'-\mathbf{k},\mathbf{G}} \\ &= \frac{1}{\Omega_c}\int_{\Omega_c} u_{n'\mathbf{k}}^*(\mathbf{r})e^{i\mathbf{G}\cdot\mathbf{r}}u_{n\mathbf{k}}(\mathbf{r})d\mathbf{r}\,\delta_{\mathbf{k}'-\mathbf{k},\mathbf{G}}. \end{aligned} \tag{35b}$$

The first-order evaluation [18] of Eq. (35b) at $\mathbf{k}' = \mathbf{k} + \mathbf{G}$ results in $\frac{1}{\Omega_c}\int_{\Omega_c} u_{n'\mathbf{k}'}^*(\mathbf{r})u_{n\mathbf{k}}(\mathbf{r})d\mathbf{r} \simeq \delta_{n',n}\delta_{\mathbf{k}',\mathbf{k}} + i\mathbf{G}\cdot\mathbf{R}_{n'n}(\mathbf{k})$, where $\mathbf{R}_{n'n}(\mathbf{k})$ is given in Eq. (18b), thus denoting transitions at the Brillouin-zone boundaries. Also $\langle m'|e^{-\hbar(\mathbf{k}'-\mathbf{k})\cdot\hat{\boldsymbol{\alpha}}_0)}|m\rangle$, with $\hat{\boldsymbol{\alpha}}_0$ of Eq. (27c), distributes the (m', m) matrix elements of $\hbar(\mathbf{k}' - \mathbf{k})\cdot\hat{\boldsymbol{\alpha}}_0$ in a noncommuting binomial distribution, which is addressed below.

In looking for the approximate solution to Eq. (33a), we first represent $|\Phi(t)\rangle$ in the same form as Eq. (17), namely,

$$\begin{aligned} |\Phi(t)\rangle &= \sum_{n',\mathbf{k}'}\sum_{m'} B_{n'\mathbf{k}'}(m', t)\psi_{n'\mathbf{K}'}(\mathbf{r}, t)|m'\rangle \\ &\quad \times e^{-\frac{i}{\hbar}\int_{t_0}^t \{\varepsilon_{n'}[\mathbf{k}'(\tau)] + E_{m'}\}d\tau}. \end{aligned} \tag{36}$$

In putting Eq. (36) into Eq. (33a), and taking the inner product using $\langle m|\psi_{n\mathbf{k}}^*$, we find after solving for $\dot{B}_{n\mathbf{k}}(m, t)$ and integration over time that

$$\begin{aligned} B_{n\mathbf{k}}(m, t) &= -\frac{1}{i\hbar}\sum_{n'\neq n}\int_{t_0}^t dt' B_{n'\mathbf{k}}(m, t') \\ &\quad \times \mathbf{F}(t')\cdot\mathbf{R}_{nn'}(\mathbf{k})e^{\frac{i}{\hbar}\int_{t_0}^{t'}[\varepsilon_n(\mathbf{k}) - \varepsilon_{n'}(\mathbf{k})]d\tau} \\ &\quad + \frac{1}{i\hbar}\sum_{n',\mathbf{k}'}\sum_{m'}\int_{t_0}^t dt' B_{n'\mathbf{k}'}(m', t')\hat{\mathcal{H}}'_{n\mathbf{k},n'\mathbf{k}'} \\ &\quad \times (m, m', t')e^{\frac{i}{\hbar}\int_{t_0}^{t'}[\varepsilon_n(\mathbf{k}) - \varepsilon_{n'}(\mathbf{k}') + E_m - E_{m'}]d\tau}. \end{aligned} \tag{37a}$$

Here, $\hat{\mathcal{H}}'$ is given by Eq. (33c), with matrix elements

$$\hat{\mathcal{H}}'_{n\mathbf{k},n'\mathbf{k}'}(m,m',t) = [V_c(\mathbf{r}+i\hbar\hat{\boldsymbol{\alpha}}_0)]_{n\mathbf{k}m,n'\mathbf{k}'m'} - \left[V_c(\mathbf{r}) + \frac{D_0^2}{\hbar\Omega}(\hat{\mathbf{p}}+\mathbf{p}_c)\cdot\boldsymbol{\lambda}^*(\hat{\mathbf{p}}+\mathbf{p}_c)\cdot\boldsymbol{\lambda}\right]_{n\mathbf{k},n'\mathbf{k}'}\delta_{m,m'} - i\hbar(\hat{\boldsymbol{\alpha}}_0)_{mm'}\cdot\dot{\mathbf{p}}_c\delta_{n,n'}\delta_{\mathbf{k},\mathbf{k}'}. \tag{37b}$$

As was done in Eq. (21a), we again seek approximate solution to Eq. (37a) in the form

$$B_{n\mathbf{k}}(m,t) \simeq \delta_{n,n_0}\delta_{\mathbf{K},\mathbf{K}_0}\delta_{m,m_0} + B^{(1)}_{n\mathbf{k}}(m,t). \tag{38a}$$

Then $B^{(1)}_{n\mathbf{k}}(m,t)$ from Eq. (37a) becomes

$$B^{(1)}_{n\mathbf{k}}(m,t) = A^{(Z)}_{n\mathbf{k}}(t) + \frac{1}{i\hbar}\int_{t_0}^{t} dt'\hat{\mathcal{H}}'_{n\mathbf{k},n_0\mathbf{k}_0}(m,m_0,t')e^{\frac{i}{\hbar}\int_{t_0}^{t'}[\varepsilon_n(\mathbf{k})-\varepsilon_{n_0}(\mathbf{k}_0)+E_m-E_{m_0}]d\tau}, \tag{38b}$$

where the matrix elements of $\hat{\mathcal{H}}'_{n\mathbf{k},n_0\mathbf{k}_0}(m,m_0,t')$ are obtained from Eq. (37b) by replacement $(n'\mathbf{k}'m')$ with $(n_0\mathbf{k}_0m_0)$, respectively; $A^{(Z)}_{n\mathbf{k}}(t)$ is defined in Eqs. (21b) and (22).

Regarding the first term in Eq. (37b), we note that, to first order in $\hat{\boldsymbol{\alpha}}_0$, assuming small magnitude of $D_0/\hbar\Omega$ (see Appendix C), $V_c(\mathbf{r}+i\hbar\hat{\boldsymbol{\alpha}}_0) = V_c(\mathbf{r}) + i\hbar\hat{\boldsymbol{\alpha}}_0\cdot\boldsymbol{\nabla}_{\mathbf{r}}V_c(\mathbf{r}) + O(\hat{\boldsymbol{\alpha}}_0^2)$. Then, the matrix elements for the first two terms in Eq. (37b) become

$$[V_c(\mathbf{r}+i\hbar\hat{\boldsymbol{\alpha}}_0)]_{mm'} - V_c(\mathbf{r})\delta_{m,m'} = -i\hbar(\hat{\boldsymbol{\alpha}}_0)_{mm'}\cdot\mathbf{F}_c(\mathbf{r}) + O(\hat{\boldsymbol{\alpha}}_0^2), \tag{39}$$

where $\mathbf{F}_c(\mathbf{r}) = -\boldsymbol{\nabla}_{\mathbf{r}}V_c(\mathbf{r})$. Then Eq. (37b) simplifies to

$$\hat{\mathcal{H}}'_{n\mathbf{k},n'\mathbf{k}'}(m,m',t) = -i\hbar(\hat{\boldsymbol{\alpha}}_0)_{mm'}\cdot[\mathbf{F}_c(\mathbf{r})_{n\mathbf{k},n'\mathbf{k}'} + \dot{\mathbf{p}}_c\delta_{n,n'}\delta_{\mathbf{k},\mathbf{k}'}] - \frac{D_0^2}{\hbar\Omega}[(\hat{\mathbf{p}}+\mathbf{p}_c)\cdot\boldsymbol{\lambda}^*(\hat{\mathbf{p}}+\mathbf{p}_c)\cdot\boldsymbol{\lambda}]_{n\mathbf{k},n'\mathbf{k}'}\delta_{m,m'}, \tag{40}$$

where $(\hat{\boldsymbol{\alpha}}_0)_{mm'}$ is now stand-alone, and $\mathbf{F}_c(\mathbf{r})$ competes directly with $\dot{\mathbf{p}}_c$. Thus, the use of Eq. (40) in Eq. (38b) greatly simplifies the approach. The calculation of $B^{(1)}_{n\mathbf{k}}(m,t)$ using Eq. (40) in Eq. (38b) is straightforward. We note that $(\hat{\boldsymbol{\alpha}}_0)_{mm_0}$ for use in Eq. (38b) is $(\hat{\boldsymbol{\alpha}}_0)_{mm_0} = (D_0/\hbar\Omega)[(m_0+1)^{1/2}\delta_{m,m_0+1}\boldsymbol{\lambda}^* - m_0^{1/2}\delta_{m,m_0-1}\boldsymbol{\lambda}]$. We then find that Eq. (38b) reduces to

$$B^{(1)}_{n\mathbf{k}}(m,t) = A^{(Z)}_{n\mathbf{k}}(t) + \frac{1}{i\hbar}\int_{t_0}^{t} dt'\left\{-i\hbar\frac{D_0}{\hbar\Omega}\left[(m_0+1)^{1/2}\delta_{m,m_0+1}\boldsymbol{\lambda}^* - m_0^{1/2}\delta_{m,m_0-1}\boldsymbol{\lambda}\right]\cdot\left[(\mathbf{F}_c)_{n\mathbf{k},n_0\mathbf{k}_0} + \dot{\mathbf{p}}_c\delta_{n,n_0}\delta_{\mathbf{k},\mathbf{k}_0}\right] - \frac{D_0^2}{\hbar\Omega}[(\hat{\mathbf{p}}+\mathbf{p}_c)\cdot\boldsymbol{\lambda}^*(\hat{\mathbf{p}}+\mathbf{p}_c)\cdot\boldsymbol{\lambda}]_{n\mathbf{k},n_0\mathbf{k}_0}\delta_{m,m_0}\right\}e^{\frac{i}{\hbar}\int_{t_0}^{t'}[\varepsilon_n(\mathbf{k})-\varepsilon_{n_0}(\mathbf{k}_0)+E_m-E_{m_0}]d\tau}. \tag{41}$$

Otherwise, we must necessarily deal with Eqs. (37a) and (37b), which are significantly more complex. In this regard, to approach the analysis of the full matrix element $\hat{\mathcal{H}}'$, appearing in Eq. (37b), we consider a complete formulation of $V_c(\mathbf{r}+i\hbar\hat{\boldsymbol{\alpha}}_0)$. Defining an operator $\hat{Q}$ such that

$$\hat{Q}V_c(\mathbf{r}) = V_c(\mathbf{r}+i\hbar\hat{\boldsymbol{\alpha}}_0), \tag{42a}$$

where

$$\hat{Q} = e^{(\hat{\mathbf{p}}+\mathbf{p}_c)\cdot\hat{\boldsymbol{\alpha}}_0} \equiv e^{\hat{U}}, \tag{42b}$$

we express $\hat{Q}$ of Eq. (42b) in the form

$$\hat{Q} = 1 + \sum_{N=1}^{\infty}\frac{\hat{U}^N}{N!}. \tag{42c}$$

Using $\hat{\boldsymbol{\alpha}}_0$ from Eq. (27c), we find that $\hat{U}$ becomes

$$\begin{aligned}\hat{U} &= \frac{D_0}{\hbar\Omega}(\hat{\mathbf{p}}+\mathbf{p}_c)\cdot(\boldsymbol{\lambda}^*\hat{a}^\dagger - \boldsymbol{\lambda}\hat{a})\\ &= \frac{D_0}{\hbar\Omega}[(\hat{\mathbf{p}}+\mathbf{p}_c)\cdot\boldsymbol{\lambda}^*\hat{a}^\dagger - (\hat{\mathbf{p}}+\mathbf{p}_c)\cdot\boldsymbol{\lambda}\hat{a}].\end{aligned} \tag{43}$$

In letting

$$\hat{A} = \varepsilon\hat{\varrho}_0^\dagger\hat{a}, \quad \hat{B} = -\varepsilon\hat{\varrho}_0\hat{a}^\dagger, \tag{44a}$$

where

$$\hat{\varrho}_0 = (\hat{\mathbf{p}}+\mathbf{p}_c)\cdot\boldsymbol{\lambda}^*, \quad \hat{\varrho}_0{}^\dagger = (\hat{\mathbf{p}}+\mathbf{p}_c)\cdot\boldsymbol{\lambda}, \tag{44b}$$

and

$$\varepsilon = \frac{D_0}{\hbar\Omega}, \tag{44c}$$

then $\hat{U}$ of Eq. (43) becomes $\hat{U} = -\varepsilon(\hat{\varrho}_0^\dagger\hat{a} - \hat{\varrho}_0\hat{a}^\dagger) = -(\hat{A}+\hat{B})$, so that

$$e^{\hat{U}} = e^{-(\hat{A}+\hat{B})} = e^{-\hat{A}}e^{-\hat{B}}e^{-\frac{1}{2}[\hat{A},\hat{B}]}. \tag{45a}$$

This follows from the use of the Baker-Campbell-Hausdorff (BCH) theorem [19], where from Eq. (44a) and the use of $[\hat{a},\hat{a}^\dagger] = 1$, it is clear that

$$[\hat{A},\hat{B}] = -\varepsilon^2\hat{\varrho}_0^\dagger\hat{\varrho}_0. \tag{45b}$$

It is noted from Eqs. (44a) and (45b) that $[\hat{A}[\hat{A},\hat{B}]] = [\hat{B}[\hat{A},\hat{B}]] = 0$, so that the use of the BCH theorem is appropriate. Therefore, it follows from Eqs. (42b) and (45a) that

$$\hat{Q} = e^{-\varepsilon\hat{\varrho}_0^\dagger\hat{a}}e^{\varepsilon\hat{\varrho}_0\hat{a}^\dagger}e^{\frac{1}{2}\varepsilon^2\hat{\varrho}_0^\dagger\hat{\varrho}_0}. \tag{46}$$

Since we seek to establish the matrix elements of $\hat{Q}V_c(\mathbf{r})$ in Eq. (42a), that is,

$$\langle\psi_{n\mathbf{K}}|\langle m|\hat{Q}V_c(\mathbf{r})|m'\rangle|\psi_{n'\mathbf{K}'}\rangle, \tag{47}$$

it is more convenient to use $\hat{Q}$ in the form of Eq. (42c). In this case, we have

$$\hat{Q} = 1 + \sum_{N=1}^{\infty}(-1)^N\frac{(\hat{A}+\hat{B})^N}{N!}, \tag{48a}$$

which is in the form of an infinite sum of binomial coefficients composed of noncommuting operators in that $[\hat{a},\hat{a}^\dagger]=1$. However, this type of binomial expression is realized through the use of the BCH theorem, namely,

$$e^{\lambda(\hat{A}+\hat{B})} = e^{\lambda\hat{A}}e^{\lambda\hat{B}}e^{-\frac{\lambda^2}{2}[\hat{A},\hat{B}]}. \tag{48b}$$

Here, the binomial expression in Eq. (48a) is obtained for the two noncommuting operators by expanding both sides of Eq. (48b) to equal orders of λ, and then comparing each side, term by term. The details of this analysis are found in Appendix D, where we find, using Eqs. (44a)–(44c), along with Eq. (D3), that

$$\frac{\hat{U}^N}{N!} = \varepsilon^N\sum_{s=0,(N-s)[2]}^{N}\frac{\left(\frac{1}{2}\hat{\varrho}_0^\dagger\hat{\varrho}_0\right)^{(N-s)/2}}{s![(N-s)/2]!} \times\sum_{r=0}^{s}(-1)^{s-r}\binom{s}{r}(\hat{\varrho}_0^\dagger\hat{a})^r(\hat{\varrho}_0\hat{a}^\dagger)^{s-r}. \tag{49}$$

In equation (49), for any integer N ranging from 1 to infinity, we note that s is in the range $[0,N]$ such that $(N-s)/2$ is an integer [this is the meaning of the indication $(N-s)[2]$].

Thus, from Eq. (42a), the term by term application on $V_c(\mathbf{r})$ gives rise to an additive N-photon process leading to the full multiphoton process. Therefore, the first term in the matrix element of $\hat{\mathcal{H}}'$ in Eq. (37b) can be determined for $[V_c(\mathbf{r}+i\hbar\hat{\boldsymbol{\alpha}}_0)]_{n\mathbf{k}m,n'\mathbf{k}'m'}$ using Eq. (49). In taking the matrix elements of (m,m') with respect to $\hat{a}^r$ and $(\hat{a}^\dagger)^{s-r}$, we find that (see Appendix E for details) matrix elements of $\hat{Q}V_c(\mathbf{r})$ become

$$\langle m'|\hat{Q}V_c(\mathbf{r})|m\rangle = \left\{\delta_{mm'} + \frac{1}{\sqrt{m'!m!}}\sum_{N=1}^{\infty}\left(\frac{D_0}{\hbar\Omega}\right)^N\sum_{s=0,(N-s)[2]}^{N}\left(\frac{1}{2}\right)^{\frac{N-s}{2}}\sum_{r=0}^{s}(m+r)!(-1)^{\frac{N-s}{2}+s-r}\binom{s}{r}(\hat{\varrho}_0^\dagger)^{\frac{N-s}{2}+r}\hat{\varrho}_0^{\frac{N+s}{2}-r}\right\}V_c(\mathbf{r}). \tag{50a}$$

Here, Eq. (50a) has been arranged in a symmetric form with respect to $\hat{\varrho}_0^m$ and $(\hat{\varrho}_0^\dagger)^m$; this will facilitate in evaluating the matrix elements with respect to $(n\mathbf{k},n'\mathbf{k}')$ over

$$\left\{\binom{s}{r}(\hat{\varrho}_0^\dagger)^{\frac{N-s}{2}+r}\hat{\varrho}_0^{\frac{N+s}{2}-r}\right\}V_c(\mathbf{r}).$$

Of course, the contribution of the order of the sum over N will be, to an extent, determined by the magnitude of $D_0/\hbar\Omega$ which is less than unity (see Appendix A). We note that with the subsequent evaluation with respect to the states $(n\mathbf{k},n'\mathbf{k}')$ over

$$\left\{\binom{s}{r}(\hat{\varrho}_0^\dagger)^{\frac{N-s}{2}+r}\hat{\varrho}_0^{\frac{N+s}{2}-r}\right\}V_c(\mathbf{r}),$$

the initial term $V_c(\mathbf{r})\delta_{m,m'}$ cancels with the counter term in Eq. (50a) of $\hat{\mathcal{H}}'$, leaving only the sum over N. Hence, as noted, each term in the sum over N is considered to be an N-photon contribution to the total $\hat{\mathcal{H}}'$ scattering process.

Thus, in fully evaluating

$$\hat{\mathcal{H}}'_{n\mathbf{k},n_0\mathbf{k}_0}(m,m_0,t')e^{\frac{i}{\hbar}\int_{t_0}^{t'}[\varepsilon_n(\mathbf{k})-\varepsilon_{n_0}(\mathbf{k}_0)+E_m-E_{m_0}]d\tau}$$

for use in Eq. (38b), it follows from Eq. (37b) that

$$\begin{aligned}\hat{\mathcal{H}}'_{n\mathbf{k},n_0\mathbf{k}_0}(m,m_0,t')e^{\frac{i}{\hbar}\int_{t_0}^{t'}[\varepsilon_n(\mathbf{k})-\varepsilon_{n_0}(\mathbf{k}_0)+E_m-E_{m_0}]d\tau} &= \left\{[V_c(\mathbf{r}+i\hbar\hat{\boldsymbol{\alpha}}_0)]_{n\mathbf{k}m,n_0\mathbf{k}_0m_0} - [V_c(\mathbf{r})]_{n\mathbf{k},n_0\mathbf{k}_0}\delta_{m,m_0}\right\}e^{\frac{i}{\hbar}\int_{t_0}^{t'}[\varepsilon_n(\mathbf{k})-\varepsilon_{n_0}(\mathbf{k}_0)+E_m-E_{m_0}]d\tau}\\ &\quad - \frac{D_0^2}{\hbar\Omega}[(\hat{\mathbf{p}}+\mathbf{p}_c)\cdot\boldsymbol{\lambda}^*(\hat{\mathbf{p}}+\mathbf{p}_c)\cdot\boldsymbol{\lambda}]_{n\mathbf{k},n_0\mathbf{k}_0}\delta_{m,m_0}e^{\frac{i}{\hbar}\int_{t_0}^{t}[\varepsilon_n(\mathbf{k})-\varepsilon_{n_0}(\mathbf{k}_0)]d\tau}\\ &\quad - i\hbar(\hat{\boldsymbol{\alpha}}_0)_{mm_0}\cdot\dot{\mathbf{p}}_c e^{\frac{i}{\hbar}\int_{t_0}^{t}(E_m-E_{m_0})d\tau}\delta_{n,n_0}\delta_{\mathbf{k},\mathbf{k}_0}.\end{aligned} \tag{50b}$$

In Eq. (50b), we note that $(\hat{\boldsymbol{\alpha}}_0)_{mm_0}$ is off-diagonal in mm_0, and the *selection rules* have been observed in the energy exponential term. Also, from Eq. (50a), it follows that

$$\begin{aligned}[V_c(\mathbf{r}+i\hbar\hat{\boldsymbol{\alpha}}_0)]_{n\mathbf{k}m,n_0\mathbf{k}_0m_0} - [V_c(\mathbf{r})]_{n\mathbf{k},n_0\mathbf{k}_0}\delta_{m,m_0} &= \frac{1}{\sqrt{m!m_0!}}\sum_{N=1}^{\infty}\left(\frac{D_0}{\hbar\Omega}\right)^N\sum_{s=0,(N-s)[2]}^{N}\left(\frac{1}{2}\right)^{\frac{N-s}{2}}\sum_{r=0}^{s}(m+r)!(-1)^{\frac{N+s}{2}-r}\binom{s}{r}\\ &\quad\times\left(\psi_{n\mathbf{k}},\left\{(\hat{\varrho}_0^\dagger)^{\frac{N-s}{2}+r}\hat{\varrho}_0^{\frac{N+s}{2}-r}V_c(\mathbf{r})\right\}\psi_{n_0\mathbf{k}_0}\right).\end{aligned} \tag{50c}$$

The specific term of Eq. (50c),

$$\Delta_{n\mathbf{k},n_0\mathbf{k}_0} = \left(\psi_{n\mathbf{k}}, \left\{(\hat{\varrho}_0^\dagger)^{\frac{N-s}{2}+r}\hat{\varrho}_0^{\frac{N+s}{2}-r}V_c(\mathbf{r})\right\}\psi_{n_0\mathbf{k}_0}\right), \tag{51}$$

can be further reduced by using the general Fourier representation for $V_c(\mathbf{r})$ of Eq. (34). Inserting $V_c(\mathbf{r})$ of Eq. (34) into Eq. (51) and noting Eq. (44b), we find that

$$\Delta_{n\mathbf{k},n_0\mathbf{k}_0} = \sum_{\mathbf{G}} U_{\mathbf{G}}[(\hbar\mathbf{G}+\mathbf{p}_c)\cdot\boldsymbol{\lambda}]^{\frac{N-s}{2}+r}[(\hbar\mathbf{G}+\mathbf{p}_c)\cdot\boldsymbol{\lambda}^*]^{\frac{N+s}{2}-r}\left(\psi_{n\mathbf{K}}, e^{i\mathbf{G}\cdot\mathbf{r}}\psi_{n_0\mathbf{K}_0}\right). \tag{52a}$$

Since

$$\left(\psi_{n\mathbf{K}}, e^{i\mathbf{G}\cdot\mathbf{r}}\psi_{n_0\mathbf{K}_0}\right) = \frac{1}{\Omega_c}\int_{\Omega_c} u^*_{n\mathbf{k}}(\mathbf{r})u_{n_0\mathbf{k}_0}(\mathbf{r})d\mathbf{r}\,\delta_{\mathbf{k}-\mathbf{k}_0,\mathbf{G}}, \tag{52b}$$

and $\mathbf{k}-\mathbf{k}_0 = \mathbf{G}$ is the selection rule emanating from the Bloch integral, we can evaluate the sum in Eq. (52a) as

$$\Delta_{n\mathbf{k},n_0\mathbf{k}_0} = U_{\mathbf{k}-\mathbf{k}_0}\{[\hbar(\mathbf{k}-\mathbf{k}_0)+\mathbf{p}_c]\cdot\boldsymbol{\lambda}\}^{\frac{N-s}{2}+r}\{[\hbar(\mathbf{k}-\mathbf{k}_0)+\mathbf{p}_c)\cdot\boldsymbol{\lambda}^*\}^{\frac{N+s}{2}-r}\frac{1}{\Omega_c}\int_{\Omega_c} u^*_{n\mathbf{k}}(\mathbf{r})u_{n_0\mathbf{k}_0}(\mathbf{r})d\mathbf{r}\,\delta_{\mathbf{k}-\mathbf{k}_0,\mathbf{G}}, \tag{52c}$$

where

$$U_{\mathbf{k}-\mathbf{k}_0} = \frac{1}{\Omega_c}\int_{\Omega_c} V_c(\mathbf{r})e^{-i(\mathbf{k}-\mathbf{k}_0)\cdot\mathbf{r}}d\mathbf{r}. \tag{52d}$$

Finally, letting $\mathbf{a} = \hbar(\mathbf{k}-\mathbf{k}_0)+\mathbf{p}_c$ and remembering from Eq. (11b) that $\boldsymbol{\lambda} = (1, i)/\sqrt{2}$, $\boldsymbol{\lambda}^* = (1, -i)/\sqrt{2}$, we find

$$\mathbf{a}\cdot\boldsymbol{\lambda} = \frac{a}{\sqrt{2}}e^{i\phi}, \quad \mathbf{a}\cdot\boldsymbol{\lambda}^* = \frac{a}{\sqrt{2}}e^{-i\phi},$$

with $a_x = a\cos\phi$ and $a_y = a\sin\phi$, and $\tan\phi = a_y/a_x$; so that $\Delta_{n\mathbf{k},n_0\mathbf{k}_0}$ of Eq. (52c) becomes

$$\Delta_{n\mathbf{k},n_0\mathbf{k}_0} = U_{\mathbf{k}-\mathbf{k}_0}O_{n\mathbf{k},n_0\mathbf{k}_0}\left(\frac{a}{\sqrt{2}}\right)^N e^{i(2r-s)\phi}e^{i\phi}, \tag{53}$$

where $U_{\mathbf{k}-\mathbf{k}_0}$ is given in Eq. (52d) and $O_{n\mathbf{k},n_0\mathbf{k}_0}$ is given in Eq. (35b). Then, inserting (53) into (50c), we obtain

$$\begin{aligned}&[V_c(\mathbf{r}+i\hbar\hat{\boldsymbol{\alpha}}_0)]_{n\mathbf{k}m,n_0\mathbf{k}_0m_0} - [V_c(\mathbf{r})]_{n\mathbf{k},n_0\mathbf{k}_0}\delta_{m,m_0}\\ &= \frac{1}{\sqrt{m!m_0!}}U_{\mathbf{k}-\mathbf{k}_0}O_{n\mathbf{k},n_0\mathbf{k}_0}\sum_{N=1}^{\infty}\left(\frac{D_0}{\hbar\Omega}\right)^N\left(\frac{a}{\sqrt{2}}\right)^N\sum_{s=0,[(N-s)/2]}^{N}\left(-\frac{1}{2}\right)^{\frac{N-s}{2}}(-1)^s\sum_{r=0}^{s}(m+r)!(-1)^r\binom{s}{r}e^{i(2r-s)\phi}.\end{aligned} \tag{54a}$$

As noted in Eq. (54a), for any integer N ranging from zero to infinity, the value of s is in the range $[0, N]$ such that $(N-s)/2$ is an integer. It then follows that for *odd values* of $N = N_o$, corresponding values of s will be odd ranging from $s_o = 1$ to N_o. As well, for *even values* of $N = N_e$, corresponding values of s will be even ranging from $s_e = 0$ to N_e. Therefore, the sum over N and s in Eq. (54a) can be expressed as

$$\sum_{N=1}^{\infty}\sum_{s=0,[(N-s)/2]}^{N} = \sum_{N_o=1}^{\infty}\sum_{s_o=1}^{N_o} + \sum_{N_e=2}^{\infty}\sum_{s_e=0}^{N_e}. \tag{54b}$$

Breaking Eq. (54a) into a sum like Eq. (54b), we let $N_o = 2N+1$ with $N = 0, 1, 2, \ldots$, and $N_e = 2N$ with $N = 1, 2, 3, \ldots$; then, $s_o = 2s+1$ and $s_e = 2s$ with s ranging from $s = 0, 1, 2, \ldots N$. Then, Eq. (54a) can be rewritten in the form of Eq. (54b) as

$$\begin{aligned}&[V_c(\mathbf{r}+i\hbar\hat{\boldsymbol{\alpha}}_0)]_{n\mathbf{k}m,n_0\mathbf{k}_0m_0} - [V_c(\mathbf{r})]_{n\mathbf{k},n_0\mathbf{k}_0}\delta_{m,m_0}\\ &= \frac{1}{\sqrt{m!m_0!}}U_{\mathbf{k}-\mathbf{k}_0}O_{n\mathbf{k},n_0\mathbf{k}_0}\left\{\sum_{N=0}^{\infty}\left(\frac{D_0}{\hbar\Omega}\right)^{2N+1}\left(\frac{a}{\sqrt{2}}\right)^{2N+1}\sum_{s_o=0}^{N}\left(-\frac{1}{2}\right)^{N-s_o}(-1)^{2s_o+1}\sum_{r=0}^{2s_o+1}(m+r)!(-1)^r\binom{2s_o+1}{r}e^{i(2r-2s_o-1)\phi}\right.\\ &\left.+\sum_{N=1}^{\infty}\left(\frac{D_0}{\hbar\Omega}\right)^{2N}\left(\frac{a}{\sqrt{2}}\right)^{2N}\sum_{s_e=0}^{N}\left(-\frac{1}{2}\right)^{N-s_e}(-1)^{2s_e}\sum_{r=0}^{2s_e}(m+r)!(-1)^r\binom{2s_e}{r}e^{i2(r-s_e)\phi}\right\}.\end{aligned} \tag{54c}$$

Equation (54c) can be further resumed and simplified. In separating the first term in the sum ($N = 1$) and then combining the remaining two terms, we find

$$\begin{aligned}&[V_c(\mathbf{r}+i\hbar\hat{\boldsymbol{\alpha}}_0)]_{n\mathbf{k}m,n_0\mathbf{k}_0m_0} - [V_c(\mathbf{r})]_{n\mathbf{k},n_0\mathbf{k}_0}\delta_{m,m_0}\\ &= \frac{1}{\sqrt{m!m_0!}}U_{\mathbf{k}-\mathbf{k}_0}O_{n\mathbf{k},n_0\mathbf{k}_0}\left\{-\frac{D_0}{\hbar\Omega}\frac{a}{\sqrt{2}}\Gamma(1)+\sum_{N=2}^{\infty}\left(\frac{D_0}{\hbar\Omega}\right)^N\left(-\frac{a}{2^{3/2}}\right)^N\sum_{s=0}^{N}(-2)^s\Gamma(s)\right\},\end{aligned} \tag{54d}$$

where

$$\Gamma(s) = \sum_{r=0}^{s} (m+r)!(-1)^r \binom{s}{r} e^{i(2r-s)\phi}. \tag{54e}$$

Here, $U_{\mathbf{k}-\mathbf{k}_0}$, $O_{n\mathbf{k},n_0\mathbf{k}_0}$, and ϕ are given in Eqs. (52d), (52b), and (53), respectively.

This establishes the exact matrix element in terms of the key relevant parameters to any desired order in $D_0/\hbar\Omega$. Thus, it follows using Eq. (50b) in Eq. (38b) that we obtain $B^{(1)}_{n\mathbf{k}}(m,t)$ for the complete formulation of $V_c(\mathbf{r}+i\hbar\hat{\boldsymbol{\alpha}}_0)$.

As a comparative note on approaches 1 and 2, in both cases, we have been working with a Hamiltonian of the form of Eq. (16a), where for both cases $\hat{H}_0$ is given in Eq. (16b); but for case 1, $\hat{H}' \equiv \hat{H}'_1$ is in Eq. (15b) and for case 2, $\hat{H}' \equiv \hat{H}'_2$ is in Eq. (33c). In case 1, given the states of $\hat{H}_0$ noted in Eq. (16b), $\hat{H}'_1$ of Eq. (15b) scatters the states of $\hat{H}_0$ to provide Zener tunneling and first-order optical absorption and emission. Whereas, in case 2, in a unitary equivalent picture, $\hat{H}'_2$ [Eq. (33c)] scatters the states of $\hat{H}_0$ from the inner crystal potential energy with $i\hbar\hat{\boldsymbol{\alpha}}_0$ serving as a spatial optical probe and coupling constant. $\hat{H}'_2$ also contains an added contribution to the perpendicular component of the kinetic energy due to the circular polarization $\boldsymbol{\lambda}$, $\boldsymbol{\lambda}^*$; as well as a contribution from the classical force $\dot{\mathbf{p}}_c$.

IV. CALCULATION OF CURRENT, WAVE FUNCTION FORMULATION

For Bloch bands, the electron current $\mathbf{j}$ is

$$\mathbf{j} = e\left\langle \frac{\hat{\mathbf{p}}}{m_e} \right\rangle = e\langle \hat{\mathbf{v}} \rangle = e \int \Phi^* \hat{\mathbf{v}} \Phi \, d\mathbf{r}, \tag{55}$$

where $\hat{\mathbf{v}}$ is the operator of the electron velocity, given by

$$\hat{\mathbf{v}} = \frac{1}{i\hbar}[\mathbf{r}, \hat{H}], \tag{56}$$

and Φ is the solution to the time-dependent Schrödinger equation.

In this section, we develop the current $\mathbf{j}$ for the two canonical Hamiltonians treated in Sec. II. Since both Hamiltonians to be treated are of the same form, i.e., $\hat{H} = \hat{H}_0 + \hat{H}'$, with identical $\hat{H}_0$ but differing $\hat{H}'$, we make use of the same duel basis set; that is, instantaneous Bloch eigenstates and harmonic-oscillator eigenstates, to represent Φ with appropriate expansion coefficients consistent with the specific time-dependent Schrödinger equation. As such, we express Φ in general as a representative solution [as in Eqs. (17) and (36)]

$$|\Phi(t)\rangle = \sum_{n,\mathbf{k}} \sum_{m} C_{n\mathbf{k}}(m,t)\psi_{n\mathbf{K}}(\mathbf{r},t)|m\rangle e^{-\frac{i}{\hbar}\int_{t_0}^{t}\{\varepsilon_n[\mathbf{k}(\tau)]+E_m\}d\tau}. \tag{57}$$

Using the wave function of Eq. (57) in $\mathbf{j}$ of Eq. (55), we find

$$\mathbf{j}(t) = e \sum_{n,\mathbf{k},m} \sum_{n',\mathbf{k}',m'} C^*_{n'\mathbf{k}'}(m',t) C_{n\mathbf{k}}(m,t) \hat{\mathbf{v}}_{n'\mathbf{k}'m',n\mathbf{k}m} e^{-\frac{i}{\hbar}\int_{t_0}^{t}[\varepsilon_n(\mathbf{k})-\varepsilon_{n'}(\mathbf{k}')+E_m-E_{m'}]d\tau}, \tag{58a}$$

where $\hat{\mathbf{v}}_{n'\mathbf{k}'m',n\mathbf{k}m}$ are matrix elements of the electron velocity given by

$$\hat{\mathbf{v}}_{n'\mathbf{k}'m',n\mathbf{k}m} = \frac{1}{i\hbar}[\mathbf{r}, \hat{\mathcal{H}}]_{n'\mathbf{k}'m',n\mathbf{k}m}. \tag{58b}$$

We note that $\mathbf{j}(t)$ of Eq. (58a) can be expressed in terms of the density matrix as

$$\mathbf{j}(t) = e \sum_{n,\mathbf{k},m} \sum_{n',\mathbf{k}',m'} \hat{\varrho}_{n\mathbf{k}m,n'\mathbf{k}'m'}(t) \hat{\mathbf{v}}_{n'\mathbf{k}'m',n\mathbf{k}m}(t),$$

so that, by comparison, protocols using the same wave functions, we see that

$$\hat{\varrho}_{n\mathbf{k}m,n'\mathbf{k}'m'}(t) = C^*_{n'\mathbf{k}'}(m',t) C_{n\mathbf{k}}(m,t) e^{-\frac{i}{\hbar}\int_{t_0}^{t}[\varepsilon_n(\mathbf{k})-\varepsilon_{n'}(\mathbf{k}')+E_m-E_{m'}]d\tau}. \tag{59}$$

While we proceed in this section by developing $C_{n\mathbf{k}}(m,t)$ to lowest order, one can use the power of density matrices in developing more advanced protocols. Such a protocol is outlined in Appendix F (density matrix formulation of current).

Now, we calculate the current for the two Hamiltonian systems addressed in Sec. II. They were generally of the form $\hat{H} = \hat{H}_0 + \hat{H}'$, where for Eqs. (16b), (15b) and for (33b), (33c), we have the following:

(1) Direct single-mode Hamiltonian

$$\hat{H}_0 = \frac{1}{2m_e}(\hat{\mathbf{p}}+\mathbf{p}_c)^2 + V_c(\mathbf{r}) + \hbar\Omega\left(\hat{a}^\dagger\hat{a} + \frac{1}{2}\right), \tag{60a}$$

$$\hat{H}'_1 = -D_0(\hat{\mathbf{p}}+\mathbf{p}_c)\cdot(\hat{a}\boldsymbol{\lambda}e^{i\mathbf{q}\cdot\mathbf{r}} + \hat{a}^\dagger\boldsymbol{\lambda}^* e^{-i\mathbf{q}\cdot\mathbf{r}}). \tag{60b}$$

(2) Glauber-transformed Hamiltonian

$$\hat{\mathcal{H}}_0 = \frac{1}{2m_e}(\hat{\mathbf{p}}+\mathbf{p}_c)^2 + V_c(\mathbf{r}) + \hbar\Omega\left(\hat{a}^\dagger\hat{a} + \frac{1}{2}\right), \tag{61a}$$

$$\hat{\mathcal{H}}'_2 = V_c(\mathbf{r}+i\hbar\hat{\boldsymbol{\alpha}}_0) - V_c(\mathbf{r}) - \frac{D_0^2}{\hbar\Omega}(\hat{\mathbf{p}}+\mathbf{p}_c)\cdot\boldsymbol{\lambda}^*\,(\hat{\mathbf{p}}+\mathbf{p}_c)\cdot\boldsymbol{\lambda} - i\hbar\hat{\boldsymbol{\alpha}}_0\cdot\dot{\mathbf{p}}_c, \tag{61b}$$

where $\hat{\boldsymbol{\alpha}}_0 = \frac{D_0}{\hbar\Omega}(\boldsymbol{\lambda}^*\hat{a}^\dagger - \boldsymbol{\lambda}\hat{a})$.

In developing $\hat{\mathbf{v}}$ of Eq. (56), the key commutation relations to be utilized are $[x_r, \hat{p}_s] = i\hbar\delta_{r,s}$ and $[\hat{a}, \hat{a}^\dagger] = 1$; all components of position and momentum commute with $\hat{a}, \hat{a}^\dagger$. $\boldsymbol{\lambda}, \boldsymbol{\lambda}^*$ are the circular polarization vectors noted below Eq. (11b); and $\mathbf{p}_c$ is a *c-number* with respect to all operators. For case 1 above, it is straightforward to show that

$$\hat{\mathbf{v}}_1 = \frac{1}{m_e}(\hat{\mathbf{p}} + \mathbf{p}_c) - D_0(\hat{a}\boldsymbol{\lambda}e^{i\mathbf{q}\cdot\mathbf{r}} + \hat{a}^\dagger\boldsymbol{\lambda}^* e^{-i\mathbf{q}\cdot\mathbf{r}}), \tag{62a}$$

and for case 2

$$\hat{\mathbf{v}}_2 = \frac{1}{m_e}(\hat{\mathbf{p}} + \mathbf{p}_c) - \frac{D_0^2}{\hbar\Omega}(\hat{\mathbf{p}} + \mathbf{p}_c)_\perp, \tag{62b}$$

where $(\hat{\mathbf{p}} + \mathbf{p}_c)_\perp = (\hat{\mathbf{p}} + \mathbf{p}_c)_x\hat{\mathbf{x}} + (\hat{\mathbf{p}} + \mathbf{p}_c)_y\hat{\mathbf{y}}$. In using the Bloch instantaneous eigenstates, $\psi_{n\mathbf{K}}$ of Eq. (8b), and the harmonic-oscillator states $(\hat{a}^\dagger\hat{a} + 1/2)|m\rangle = E_m|m\rangle$, where $E_m = \hbar\Omega(m + 1/2)$, $m = 0, 1, 2, \ldots$, we find

$$(\hat{\mathbf{v}}_1)_{n'\mathbf{k}'m',n\mathbf{k}m} = \frac{1}{m_e}(\hat{\mathbf{p}}_{n'n} + \mathbf{p}_c\delta_{n'n})\delta_{\mathbf{k}',\mathbf{k}}\delta_{m'm} - D_0(\boldsymbol{\lambda}\hat{a}_{m'm}\delta_{\mathbf{k}',\mathbf{k}+\mathbf{q}} + \boldsymbol{\lambda}^*\hat{a}^\dagger_{m'm}\delta_{\mathbf{k}',\mathbf{k}-\mathbf{q}}). \tag{63a}$$

Here, $\hat{a}_{m'm} = m^{1/2}\delta_{m',m-1}$, $\hat{a}^\dagger_{m'm} = (m+1)^{1/2}\delta_{m',m+1}$, and the terms $\delta_{\mathbf{k}',\mathbf{k}\pm\mathbf{q}}$ come from the radial contribution from the matrix elements. Also, we find

$$(\hat{\mathbf{v}}_2)_{n'\mathbf{k}'m',n\mathbf{k}m} = \left[\frac{1}{m_e}(\hat{\mathbf{p}}_{n'n} + \mathbf{p}_c\delta_{n'n}) - \frac{D_0^2}{\hbar\Omega}(\hat{\mathbf{p}}_{n'n} + \mathbf{p}_c\delta_{n'n})_\perp\right]\delta_{\mathbf{k}',\mathbf{k}}\delta_{m',m}. \tag{63b}$$

Inserting the matrix elements of $\hat{\mathbf{v}}_1$ and $\hat{\mathbf{v}}_2$ from Eqs. (63a) and (63b) into the current expression of Eq. (58a), we find that

$$\begin{aligned}\mathbf{j}_1(t) = e\sum_{n,\mathbf{k},m}\sum_{n'}\Big\{&C^*_{n'\mathbf{k}}(m,t)C_{n\mathbf{k}}(m,t)\frac{1}{m_e}(\hat{\mathbf{p}}_{n'n} + \mathbf{p}_c\delta_{n'n})e^{-\frac{i}{\hbar}\int_{t_0}^t[\varepsilon_n(\mathbf{k})-\varepsilon_{n'}(\mathbf{k})]d\tau}\\ &- D_0\big[C^*_{n'\mathbf{k}+\mathbf{q}}(m-1,t)C_{n\mathbf{k}}(m,t)m^{1/2}\boldsymbol{\lambda}e^{-\frac{i}{\hbar}\int_{t_0}^t[\varepsilon_n(\mathbf{k})-\varepsilon_{n'}(\mathbf{k}+\mathbf{q})+\hbar\Omega]d\tau}\\ &+ C^*_{n'\mathbf{k}-\mathbf{q}}(m+1,t)C_{n\mathbf{k}}(m,t)(m+1)^{1/2}\boldsymbol{\lambda}^*e^{-\frac{i}{\hbar}\int_{t_0}^t[\varepsilon_n(\mathbf{k})-\varepsilon_{n'}(\mathbf{k}-\mathbf{q})-\hbar\Omega]d\tau}\big]\Big\},\end{aligned} \tag{64a}$$

and

$$\mathbf{j}_2(t) = e\sum_{n,n'}\sum_m C^*_{n'\mathbf{k}}(m,t)C_{n\mathbf{k}}(m,t)\left[\frac{1}{m_e}(\hat{\mathbf{p}}_{n'n} + \mathbf{p}_c\delta_{n'n}) - \frac{D_0^2}{\hbar\Omega}(\hat{\mathbf{p}}_{n'n} + \mathbf{p}_c\delta_{n'n})_\perp\right]e^{-\frac{i}{\hbar}\int_{t_0}^t[\varepsilon_n(\mathbf{k})-\varepsilon_{n'}(\mathbf{k})]d\tau}. \tag{64b}$$

We generally observe that $\mathbf{j}_1(t)$ is an implicit function of $(n\mathbf{k}m)$ whereas $\mathbf{j}_2(t)$ is dependent upon only $(n\mathbf{k})$ since the sum $\sum_m C^*_{n'\mathbf{k}}(m,t)C_{n\mathbf{k}}(m,t) \equiv F_{n'\mathbf{k}n\mathbf{k}}(t)$. This suggests that the unitary transformation to the canonical form (case 2) may be quite fruitful for studying the spectral content of the Bloch bands from frequency-dependent Fourier analysis of $\mathbf{j}_2(t)$.

Next, in determining $C_{n\mathbf{k}}(m,t)$, we put $\Phi(t)$ of Eq. (57) into the Schrödinger equation (7) and take the scalar products with respect to $\psi_{n\mathbf{K}}(\mathbf{r},t)|m\rangle$; using the orthogonality properties of both $\psi_{n\mathbf{K}}(\mathbf{r},t)$ and $|m\rangle$, we obtain

$$\begin{aligned}\dot{C}_{n\mathbf{k}}(m,t) = &-\frac{1}{i\hbar}\sum_{n'\neq n}C_{n'\mathbf{k}}(m,t)\mathbf{F}(t)\cdot\mathbf{R}_{nn'}(\mathbf{k})e^{\frac{i}{\hbar}\int_{t_0}^t[\varepsilon_n(\mathbf{k})-\varepsilon_{n'}(\mathbf{k})]d\tau}\\ &+\frac{1}{i\hbar}\sum_{n'\mathbf{k}'}\sum_{m'}C_{n'\mathbf{k}'}(m',t)\hat{\mathcal{H}}'_{n\mathbf{k},n'\mathbf{k}'}(m,m',t)e^{\frac{i}{\hbar}\int_{t_0}^t[\varepsilon_n(\mathbf{k})-\varepsilon_{n'}(\mathbf{k}')+E_m-E_{m'}]d\tau},\end{aligned} \tag{65}$$

where $\mathbf{F}(t)$ and $\mathbf{R}_{nn'}(\mathbf{k})$ are given in Eq. (18b), and $\hat{\mathcal{H}}'$ corresponds to either Eq. (15b) or Eq. (33c) for this current calculation. From Eq. (65), it follows after integration

$$\begin{aligned}C_{n\mathbf{k}}(m,t) = &-\frac{1}{i\hbar}\sum_{n'\neq n}\int_{t_0}^t dt' C_{n'\mathbf{k}}(m,t')\mathbf{F}(t')\cdot\mathbf{R}_{nn'}(\mathbf{k})e^{\frac{i}{\hbar}\int_{t_0}^{t'}[\varepsilon_n(\mathbf{k})-\varepsilon_{n'}(\mathbf{k})]d\tau}\\ &+\frac{1}{i\hbar}\sum_{n'\mathbf{k}'}\sum_{m'}\int_{t_0}^t dt' C_{n'\mathbf{k}'}(m',t')\hat{\mathcal{H}}'_{n\mathbf{k},n'\mathbf{k}'}(m,m',t')e^{\frac{i}{\hbar}\int_{t_0}^{t'}[\varepsilon_n(\mathbf{k})-\varepsilon_{n'}(\mathbf{k}')+E_m-E_{m'}]d\tau}.\end{aligned} \tag{66}$$

Seeking an approximate solution [an alternative solution to Eq. (65) is discussed in Appendix G] to Eq. (66) in the form

$$C_{n\mathbf{k}}(m,t) \simeq \delta_{n,n_0}\delta_{\mathbf{K},\mathbf{K}_0}\delta_{m,m_0} + C^{(1)}_{n\mathbf{k}}(m,t), \tag{67a}$$

where $(n_0\mathbf{K}_0m_0)$ are initial states of the system, that is, $C_{n\mathbf{k}}(m,t_0)$ [20], we find, after insertion into the right-hand side of Eq. (66), the first-order solution for $C_{n\mathbf{k}}(m,t)$ to be

$$C^{(1)}_{n\mathbf{k}}(m,t) \simeq -\frac{1}{i\hbar}\int_{t_0}^{t} dt'\mathbf{F}(t')\cdot\mathbf{R}_{nn_0}(\mathbf{k}_0)e^{\frac{i}{\hbar}\int_{t_0}^{t'}[\varepsilon_n(\mathbf{k}_0)-\varepsilon_{n_0}(\mathbf{k}_0)]d\tau}\delta_{\mathbf{k},\mathbf{k}_0}\delta_{m,m_0}$$
$$+\frac{1}{i\hbar}\int_{t_0}^{t} dt'\hat{\mathcal{H}}'_{n\mathbf{k},n_0\mathbf{k}_0}(m,m_0,t')e^{\frac{i}{\hbar}\int_{t_0}^{t'}[\varepsilon_n(\mathbf{k})-\varepsilon_{n_0}(\mathbf{k}_0)+E_m-E_{m_0}]d\tau}. \tag{67b}$$

In $C^{(1)}_{n\mathbf{k}}(m,t)$ of Eq. (67b), the first term describes the classic Zener tunneling component of the transition probability, noted by $A^{(Z)}_{n\mathbf{k}}(m,t)$ in Eqs. (21b) and (22), whereas the second term represents the first-order transitional behavior due to $\hat{\mathcal{H}}'$.

In putting Eq. (67a) into $\mathbf{j}(t)$ of Eq. (58a) and keeping terms to first order in $C^{(1)}_{n\mathbf{k}}$, we find

$$\mathbf{j}(t) = e\left[\hat{\mathbf{v}}_{n_0\mathbf{k}_0m_0,n_0\mathbf{k}_0m_0} + 2\mathrm{Re}\sum_{n,\mathbf{k}}\sum_{m} C^{(1)}_{n\mathbf{k}}(m,t)\hat{\mathbf{v}}_{n_0\mathbf{k}_0m_0,n\mathbf{k}m}(t)e^{-\frac{i}{\hbar}\int_{t_0}^{t}[\varepsilon_n(\mathbf{k})-\varepsilon_{n_0}(\mathbf{k}_0)+E_m-E_{m_0}]d\tau}\right]. \tag{68}$$

The results in $\mathbf{j}(t)$ of Eq. (68) will, of course, depend on the behavior of $C^{(1)}_{n\mathbf{k}}(m,t)$, which is determined by Eq. (67b) for a given $\hat{\mathcal{H}}'$; and will depend on the matrix elements of the velocity, $\mathbf{v}_{n_0\mathbf{k}_0m_0,n\mathbf{k}m}$, given in Eqs. (63a) and (63b) depending on the transformation we explore. It then follows that by inserting $C^{(1)}_{n\mathbf{k}}(m,t)$ of Eq. (67b) into $\mathbf{j}(t)$ of Eq. (68), we obtain to first order

$$\mathbf{j}(t) = e\Bigg[\hat{\mathbf{v}}_{n_0\mathbf{k}_0m_0,n_0\mathbf{k}_0m_0}(t) + 2\mathrm{Re}\sum_{n\neq n_0} A^{(Z)}_{n\mathbf{k}_0}(m_0,t)\hat{\mathbf{v}}_{n_0\mathbf{k}_0m_0,n\mathbf{k}_0m_0}(t)e^{-\frac{i}{\hbar}\int_{t_0}^{t}[\varepsilon_n(\mathbf{k}_0)-\varepsilon_{n_0}(\mathbf{k}_0)]d\tau}$$
$$+2\mathrm{Re}\sum_{n,\mathbf{k}}\sum_{m}\frac{1}{i\hbar}\int_{t_0}^{t} dt'\hat{\mathcal{H}}'_{n\mathbf{k},n_0\mathbf{k}_0}(m,m_0,t')\hat{\mathbf{v}}_{n_0\mathbf{k}_0m_0,n\mathbf{k}m}(t)e^{-\frac{i}{\hbar}\int_{t'}^{t}[\varepsilon_n(\mathbf{k})-\varepsilon_{n_0}(\mathbf{k}_0)+E_m-E_{m_0}]d\tau}\Bigg]. \tag{69a}$$

In Eq. (69a), the velocity matrix elements in the two first terms are determined from Eqs. (63a) and (63b) for each case 1 or 2 considered. In all situations,

$$\hat{\mathbf{p}}_{n_0,n_0}(\mathbf{k}) = \frac{m_e}{\hbar}\nabla_{\mathbf{k}}\varepsilon_{n_0}(\mathbf{k}), \tag{69b}$$

and

$$\hat{\mathbf{p}}_{n_0,n}(\mathbf{k}) = \frac{1}{\Omega_c}\int_{\Omega_c} u^*_{n_0\mathbf{k}}(\mathbf{r})\hat{\mathbf{p}}u_{n\mathbf{k}}(\mathbf{r})d\mathbf{r} = \frac{m_e}{i\hbar}[\varepsilon_n(\mathbf{k})-\varepsilon_{n_0}(\mathbf{k})]\mathbf{R}_{n_0n}(\mathbf{k}). \tag{69c}$$

The results for the third term are determined by inserted both matrix elements for $\hat{\mathcal{H}}'$ and $\hat{\mathbf{v}}$ into the expression and evaluating the sum over $(n\mathbf{k}m)$ while observing the implicit selection rules present in the matrix elements.

For case 1, with $\hat{\mathcal{H}}'$ given by Eq. (60b) and $\hat{\mathbf{v}}_1$ by Eq. (62a), we find that

$$\hat{\mathcal{H}}'_{n\mathbf{k},n_0\mathbf{k}_0}(m,m_0,t) \equiv \big(\psi_{n\mathbf{K}},\langle m|\hat{\mathcal{H}}'|m_0\rangle\psi_{n_0\mathbf{K}_0}\big)$$
$$= -D_0\big[m_0^{1/2}(\hat{\mathbf{p}}+\mathbf{p}_c)_{nn_0}\cdot\boldsymbol{\lambda}\delta_{\mathbf{k},\mathbf{k}_0+\mathbf{q}}\delta_{m,m_0-1} + (m_0+1)^{1/2}(\hat{\mathbf{p}}+\mathbf{p}_c)_{nn_0}\cdot\boldsymbol{\lambda}^*\delta_{\mathbf{k},\mathbf{k}_0-\mathbf{q}}\delta_{m,m_0+1}\big], \tag{70a}$$

and

$$(\hat{\mathbf{v}}_1)_{n_0\mathbf{k}_0m_0,n\mathbf{k}m} = \frac{1}{m_e}(\hat{\mathbf{p}}+\mathbf{p}_c)_{n_0n}\delta_{\mathbf{k}_0,\mathbf{k}}\delta_{m_0,m} - D_0\big[m^{1/2}\boldsymbol{\lambda}\delta_{\mathbf{k}_0,\mathbf{k}+\mathbf{q}}\delta_{m_0,m-1} + (m+1)^{1/2}\boldsymbol{\lambda}^*\delta_{\mathbf{k}_0,\mathbf{k}-\mathbf{q}}\delta_{m_0,m+1}\big]\delta_{n,n_0}. \tag{70b}$$

Putting the matrix elements of Eqs. (70a) and (70b) into $\mathbf{j}(t)$ of Eq. (69a), we find after combining terms that, for case 1,

$$\mathbf{j}_1(t) = e\Bigg\{\frac{1}{\hbar}\nabla_{\mathbf{k}}\varepsilon_{n_0}(\mathbf{k})|_{\mathbf{k}=\mathbf{k}_0} + 2\mathrm{Re}\sum_{n\neq n_0} A^{(Z)}_{n\mathbf{k}_0}(m_0,t)\frac{1}{m_e}\hat{\mathbf{p}}_{nn_0}(\mathbf{k}_0)e^{-\frac{i}{\hbar}\int_{t_0}^{t}[\varepsilon_n(\mathbf{k}_0)-\varepsilon_{n_0}(\mathbf{k}_0)]d\tau}$$
$$+2D_0^2\mathrm{Re}\frac{1}{i\hbar}\sum_{n}\Bigg[m_0\int_{t_0}^{t} dt'\boldsymbol{\lambda}^*(\hat{\mathbf{p}}+\mathbf{p}_c)_{nn_0}\cdot\boldsymbol{\lambda}e^{-\frac{i}{\hbar}\int_{t'}^{t}[\varepsilon_n(\mathbf{k}_0+\mathbf{q})-\varepsilon_{n_0}(\mathbf{k}_0)-\hbar\Omega]d\tau}$$
$$+(m_0+1)\int_{t_0}^{t} dt'\boldsymbol{\lambda}(\hat{\mathbf{p}}+\mathbf{p}_c)_{nn_0}\cdot\boldsymbol{\lambda}^*e^{-\frac{i}{\hbar}\int_{t'}^{t}[\varepsilon_n(\mathbf{k}_0-\mathbf{q})-\varepsilon_{n_0}(\mathbf{k}_0)+\hbar\Omega]d\tau}\Bigg]\Bigg\}. \tag{71}$$

For case 2, $\hat{\mathcal{H}}_2'$ is given by Eq. (61b) and $\hat{\mathbf{v}}_2$ is given by Eq. (62b). In considering the case when $\hat{\boldsymbol{\alpha}}_0$ is small, we can expand the key component of $\hat{\mathcal{H}}_2'$ so that it is represented in the form

$$\hat{\mathcal{H}}_2' \simeq -i\hbar\hat{\boldsymbol{\alpha}}_0 \cdot (\mathbf{F}_c + \dot{\mathbf{p}}_c) - \frac{D_0^2}{\hbar\Omega}(\hat{\mathbf{p}} + \mathbf{p}_c) \cdot \boldsymbol{\lambda}^* (\hat{\mathbf{p}} + \mathbf{p}_c) \cdot \boldsymbol{\lambda}. \tag{72a}$$

Then, the key matrix element is

$$(\hat{\mathcal{H}}_2')_{n\mathbf{k},n_0\mathbf{k}_0}(m, m_0, t) \simeq -i\hbar(\hat{\boldsymbol{\alpha}}_0)_{mm_0} \cdot \left[(\mathbf{F}_c)_{n\mathbf{k},n_0\mathbf{k}_0} + \dot{\mathbf{p}}_c\delta_{n,n_0}\delta_{\mathbf{k},\mathbf{k}_0}\right] - \frac{D_0^2}{\hbar\Omega}[(\hat{\mathbf{p}} + \mathbf{p}_c) \cdot \boldsymbol{\lambda}^* (\hat{\mathbf{p}} + \mathbf{p}_c) \cdot \boldsymbol{\lambda}]_{n\mathbf{k},n_0\mathbf{k}_0}\delta_{m,m_0}, \tag{72b}$$

where $(\hat{\boldsymbol{\alpha}}_0)_{mm_0}$ is given below Eq. (40). As well, as noted in Eq. (63b),

$$(\hat{\mathbf{v}}_2)_{n_0\mathbf{k}_0m_0,n\mathbf{k}m} = \left[\frac{1}{m_e}(\hat{\mathbf{p}}_{n_0n} + \mathbf{p}_c\delta_{n_0,n}) - \frac{D_0^2}{\hbar\Omega}(\hat{\mathbf{p}}_{n_0n} + \mathbf{p}_c\delta_{n_0,n})_\perp\right]\delta_{\mathbf{k}_0,\mathbf{k}}\delta_{m_0,m}. \tag{72c}$$

The strength of the perpendicular component in $\hat{\mathbf{v}}_2$ of Eq. (72c) is easily noted by adapting to the parametrization of Appendix C so that $m_eD_0^2/\hbar\Omega = 1/(1+2\omega^2/\omega_p^2)$. Then, in choosing $\omega_p \sim 10^{16}$ rad/s (metallic-like), we estimate that $m_eD_0^2/\hbar\Omega \ll 1$ for frequencies above the UV. Whereas, for $\omega_p \sim 10^{14}$ rad/s (a heavily doped semiconductor), $m_eD_0^2/\hbar\Omega$ remains small for frequencies above the far IR. Thus, the contributing strength of $m_eD_0^2/\hbar\Omega$ is determined by the density of free carriers provided by the specific material system.

Then, using the matrix elements of $(\hat{\mathcal{H}}_2')_{n\mathbf{k},n_0\mathbf{k}_0}(m, m_0, t)$ and $(\hat{\mathbf{v}}_2)_{n_0\mathbf{k}_0m_0,n\mathbf{k}m}$ in $\mathbf{j}(t)$ of Eq. (69a), we find for case 2 that

$$\begin{aligned}\mathbf{j}_2(t) = e\Bigg\{&\frac{1}{m_e}\big[\hat{\mathbf{p}}_{n_0n_0}(\mathbf{k}_0) + \mathbf{p}_c\big] - \frac{D_0^2}{\hbar\Omega}\big[\hat{\mathbf{p}}_{n_0n_0}(\mathbf{k}_0) + \mathbf{p}_c\big]_\perp \\ &+ 2\mathrm{Re}\sum_{n\neq n_0} A_{n\mathbf{k}_0}^{(Z)}(m_0, t)\left[\frac{1}{m_e}\hat{\mathbf{p}}_{n_0n}(\mathbf{k}_0) - \frac{D_0^2}{\hbar\Omega}\hat{\mathbf{p}}_{\perp n_0n}(\mathbf{k}_0)\right]e^{-\frac{i}{\hbar}\int_{t_0}^t[\varepsilon_n(\mathbf{k}_0)-\varepsilon_{n_0}(\mathbf{k}_0)]d\tau} \\ &- 2\frac{D_0^2}{\hbar\Omega}\mathrm{Re}\sum_n \frac{1}{i\hbar}\int_{t_0}^t dt'[(\hat{\mathbf{p}}+\mathbf{p}_c)\cdot\boldsymbol{\lambda}^* (\hat{\mathbf{p}}+\mathbf{p}_c)\cdot\boldsymbol{\lambda}]_{n\mathbf{k}_0,n_0\mathbf{k}_0} \\ &\times\left[\frac{1}{m_e}(\hat{\mathbf{p}}_{n_0n} + \mathbf{p}_c\delta_{n_0,n}) - \frac{D_0^2}{\hbar\Omega}(\hat{\mathbf{p}}_{n_0n} + \mathbf{p}_c\delta_{n_0,n})_\perp\right]_{\mathbf{k}=\mathbf{k}_0(t)} e^{-\frac{i}{\hbar}\int_{t'}^t[\varepsilon_n(\mathbf{k}_0)-\varepsilon_{n_0}(\mathbf{k}_0)]d\tau}\Bigg\}.\end{aligned} \tag{73}$$

We note in Eq. (73) that

$$\frac{1}{m_e}(\hat{\mathbf{p}} + \mathbf{p}_c) - \frac{D_0^2}{\hbar\Omega}(\hat{\mathbf{p}} + \mathbf{p}_c)_\perp$$

can be expressed as

$$\frac{1}{m_e}\left[\left(1 - \frac{m_eD_0^2}{\hbar\Omega}\right)(\hat{\mathbf{p}} + \mathbf{p}_c)_\perp\right] + \frac{1}{m_e}(\hat{\mathbf{p}} + \mathbf{p}_c)_z;$$

here,

$$1 - \frac{m_eD_0^2}{\hbar\Omega} = \frac{2\omega^2/\omega_p^2}{1 + 2\omega^2/\omega_p^2}.$$

This shows that low-frequency quantum radiation values quench the perpendicular component of $(\hat{\mathbf{p}} + \mathbf{p}_c)$, whereas high-frequency values tend to restore the fractional coefficient of $(\hat{\mathbf{p}} + \mathbf{p}_c)$ to unity.

Finally, with regard to case 2, should one want to consider the evaluation of $\mathbf{j}(t)$ in Eq. (69a) for arbitrary $\hat{\boldsymbol{\alpha}}_0$, then we would need to use $\hat{\mathcal{H}}'_{n\mathbf{k},n_0\mathbf{k}_0}(m, m_0, t')e^{\frac{i}{\hbar}\int_{t_0}^{t'}[\varepsilon_n(\mathbf{k})-\varepsilon_{n_0}(\mathbf{k}_0)+E_m-E_{m_0}]d\tau}$ derived from Eq. (50b). In the calculations, $\hat{\mathbf{v}}_2$ for use in Eq. (69a) is given in Eq. (63b) as used for the case of $\hat{\boldsymbol{\alpha}}_0$ small. Also, $\hat{\varrho}_0$ and $\hat{\varrho}_0^\dagger$ are specified in Eq. (44b). We then find that $\mathbf{j}_2(t)$ becomes

$$\begin{aligned}\mathbf{j}_2(t) = e\Bigg\{&\frac{1}{m_e}\big[\hat{\mathbf{p}}_{n_0n_0}(\mathbf{k}_0) + \mathbf{p}_c\big] - \frac{D_0^2}{\hbar\Omega}\big[\hat{\mathbf{p}}_{n_0n_0}(\mathbf{k}_0) + \mathbf{p}_c\big]_\perp \\ &+ 2\mathrm{Re}\sum_{n\neq n_0} A_{n\mathbf{k}_0}^{(Z)}(m_0, t)\left[\frac{1}{m_e}\hat{\mathbf{p}}_{n_0n}(\mathbf{k}_0) - \frac{D_0^2}{\hbar\Omega}\hat{\mathbf{p}}_{\perp n_0n}(\mathbf{k}_0)\right]e^{-\frac{i}{\hbar}\int_{t_0}^t[\varepsilon_n(\mathbf{k}_0)-\varepsilon_{n_0}(\mathbf{k}_0)]d\tau} \\ &+ 2\mathrm{Re}\sum_n \frac{1}{i\hbar}\int_{t_0}^t dt'\Big\{[V_c(\mathbf{r} + i\hbar\hat{\boldsymbol{\alpha}}_0)]_{n\mathbf{k}_0m_0,n_0\mathbf{k}_0m_0} - [V_c(\mathbf{r})]_{n\mathbf{k}_0,n_0\mathbf{k}_0}\end{aligned}$$

$$- \frac{D_0^2}{\hbar\Omega}[(\hat{\mathbf{p}}+\mathbf{p}_c)\cdot\boldsymbol{\lambda}^*(\hat{\mathbf{p}}+\mathbf{p}_c)\cdot\boldsymbol{\lambda}]_{n\mathbf{k}_0,n_0\mathbf{k}_0}\bigg\}$$

$$\times\left[\frac{1}{m_e}(\hat{\mathbf{p}}_{n_0 n}+\mathbf{p}_c\delta_{n_0,n}) - \frac{D_0^2}{\hbar\Omega}(\hat{\mathbf{p}}_{n_0 n}+\mathbf{p}_c\delta_{n_0,n})_\perp\right]_{\mathbf{k}=\mathbf{k}_0(t)} e^{-\frac{i}{\hbar}\int_{t'}^{t}[\varepsilon_n(\mathbf{k}_0)-\varepsilon_{n_0}(\mathbf{k}_0)]d\tau}\Bigg\}. \quad (74)$$

Here, the $[V_c(\mathbf{r}+i\hbar\hat{\boldsymbol{\alpha}}_0)-V_c(\mathbf{r})]$ matrix elements are given in Eq. (54d). $\mathbf{j}_2(t)$ of Eqs. (73) and (74) is rich in spectral information relevant to the Bloch bands. A major part of the temporal dependence of Eq. (74) is denoted in the third term of $\mathbf{j}_2(t)$ by [we note that the other matrix element terms are also time dependent through $\mathbf{p}_c(t)$]

$$I_{nn_0}(t) = \int_{t_0}^{t} dt' e^{-\frac{i}{\hbar}\int_{t'}^{t}[\varepsilon_n(\mathbf{k}_0)-\varepsilon_{n_0}(\mathbf{k}_0)]d\tau}; \quad (75a)$$

here,

$$\varepsilon_n(\mathbf{k}) = \sum_{\mathbf{l}} \varepsilon_n(\mathbf{l}) e^{i\mathbf{k}\mathbf{l}} e^{i\mathbf{k}_c\mathbf{l}}, \quad (75b)$$

where $\mathbf{k}_c(t) = \mathbf{p}_c(t)/\hbar$.

The Fourier transform of $\mathbf{j}_2(t)$ and $I(t)$ aids in revealing the detailed nature of the spectral structure of ε_n with respect to ε_{n_0}. $\varepsilon_n(\mathbf{k})$ is mediated by the classical field which can be used as a tuning probe. But for simplicity here, we set $\mathbf{p}_c = 0$ and $t_0 = 0$ in $\mathbf{j}_2(t)$, and we find that

$$I_{nn_0}(t) = i\hbar\frac{e^{-\frac{i}{\hbar}[\varepsilon_n(\mathbf{K}_0)-\varepsilon_{n_0}(\mathbf{K}_0)]t}-1}{\varepsilon_n(\mathbf{K}_0)-\varepsilon_{n_0}(\mathbf{K}_0)}. \quad (76)$$

Here, $\varepsilon_n(\mathbf{K}_0)-\varepsilon_{n_0}(\mathbf{K}_0)$ is the energy spacing between bands n_0 and n at the Brillouin-zone value $\mathbf{K}_0$. $\mathbf{j}(t)$ has a frequency-dependent Fourier transform which can be evaluated with the use of a δ function plus a principal value [21] highlighting the state sums over energy differences; that is, through the use of

$$\int_0^\infty e^{iKx}dK = i\frac{\mathrm{P}}{x} + \pi\delta(x).$$

A complete spectral intensity analysis of $\mathbf{j}_1(t)$ and $\mathbf{j}_2(t)$ would entail the evaluation of $S_j(\omega) \sim \omega^2|\mathbf{j}(\omega)|^2$ [22]; such a complex numerical analysis is not intended in this work.

In the overall analysis of the current, note that $(\hat{\mathbf{v}}_1)_{n'\mathbf{k}'m',n\mathbf{k}m}$ of Eq. (63a) is off-diagonal in (m, m'), whereas $(\hat{\mathbf{v}}_2)_{n'\mathbf{k}'m',n\mathbf{k}m}$ of Eq. (63b) is diagonal in (m, m'); this results in the current $\mathbf{j}_1(t)$ of Eq. (71) possessing an explicit optical absorption or emission component. Whereas in $\mathbf{j}_2(t)$ of Eqs. (73) and (74), the current expression depends solely on $(n\mathbf{k})$. This is observed generally in Eq. (64b).

V. SUMMARY AND CONCLUSIONS

Electron dynamics has been developed for a Bloch electron accelerating in a homogeneous external electric field of arbitrary time dependence while interacting with a quantized electromagnetic radiation field. In considering the single-mode description for the free-space quantized radiation field in circular polarization state, we find, using as basis the instantaneous Bloch and harmonic-oscillator eigenstates, that the first-order solution to the time-dependent Schrödinger equation yields Zener tunneling along with direct single-photon optical absorption and emission.

In a canonical scenario, utilizing a Glauber-like displacement unitary transformation of the initial problem, we see a scenario which is significantly more revealing of the solid-state spectral character as reflected in the Bloch electron current. Moreover, the canonical form introduced a spatial *quantum spectral* probative nature of the inner crystal potential energy through $i\hbar\hat{\boldsymbol{\alpha}}_0$, which, when fully decomposed in the transformed Hamiltonian, revealed the full spectral content of the Bloch Hamiltonian even through the lowest-order calculated Bloch electron current.

APPENDIX A: SOLID-STATE DIPOLE APPROXIMATION

The central perturbing Hamiltonian for the single-mode problem is noted in Eq. (15b) as

$$\hat{H}' = -D_0(\hat{\mathbf{p}}+\mathbf{p}_c)\cdot(\hat{a}\boldsymbol{\lambda}e^{i\mathbf{q}\cdot\mathbf{r}} + \hat{a}^\dagger\boldsymbol{\lambda}^* e^{-i\mathbf{q}\cdot\mathbf{r}}). \quad (A1)$$

Since $\mathbf{r}$ and $\hat{\mathbf{p}}$ do not commute with each other, but do commute with $\hat{a}$ and $\hat{a}^\dagger$, we take the matrix elements of $\hat{H}'$ with respect to the Bloch instantaneous eigenstates of Eq. (8a). Then, we find

$$(\hat{H}')_{n'\mathbf{k}',n\mathbf{k}} = -D_0(\hat{a}\hat{S}_{n'\mathbf{k}',n\mathbf{k}} + \hat{a}^\dagger\hat{S}^\dagger_{n'\mathbf{k}',n\mathbf{k}}), \quad (A2)$$

where

$$\hat{S}_{n'\mathbf{k}',n\mathbf{k}} = \int_V \psi^*_{n'\mathbf{K}'}(\mathbf{r})(\hat{\mathbf{p}}+\mathbf{p}_c)\cdot\boldsymbol{\lambda}e^{i\mathbf{q}\cdot\mathbf{r}}\psi_{n\mathbf{K}}(\mathbf{r})d\mathbf{r}$$
$$= \frac{1}{\Omega_c}\int_{\Omega_c} u^*_{n'\mathbf{k}'}(\mathbf{r})(\hat{\mathbf{p}}+\mathbf{p}_c)\cdot\boldsymbol{\lambda}u_{n\mathbf{k}}(\mathbf{r})d\mathbf{r}\delta_{\mathbf{k}',\mathbf{k}+\mathbf{q}+\mathbf{G}}, \quad (A3)$$

and

$$\hat{S}^\dagger_{n'\mathbf{k}',n\mathbf{k}} = \frac{1}{\Omega_c}\int_{\Omega_c} u^*_{n'\mathbf{k}'}(\mathbf{r})(\hat{\mathbf{p}}+\mathbf{p}_c)\cdot\boldsymbol{\lambda}^* u_{n\mathbf{k}}(\mathbf{r})d\mathbf{r}\delta_{\mathbf{k}',\mathbf{k}-\mathbf{q}+\mathbf{G}}. \quad (A4)$$

In considering the first Brillouin zone, we take $\mathbf{G} = 0$ so that $\mathbf{k}' = \mathbf{k} \pm \mathbf{q}$ in Eqs. (A3) and (A4).

We note that in estimating the "order of magnitude" $\mathbf{k}$ value as $2\pi/a_L$ with $a_L = 5\times10^{-8}$ cm, an approximate lattice parameter, and using $q = \omega/c$ with $c = 3\times10^{10}$ cm/s for the momentum of photons, we find that k/q is much greater than unity for a frequency range from less than microwaves into the deep UV, greater than unity for frequencies ranging into the x-ray region, but less than unity for γ rays and beyond. Thus, for Eqs. (A1) and (A2) in the analysis, we adopt $k \gg q$, so that on the scale of a lattice parameter the solid-state dipole approximation is in effect. It is then clear from Eqs. (A1) and (A2) that $(|\mathbf{k}|, |\mathbf{k}'|) \gg |\mathbf{q}|$, so that $e^{i\mathbf{q}\cdot\mathbf{r}}$ can be suppressed in the matrix elements of $\hat{H}'$.

A further, more quantitative consideration concerning the magnitude of $\mathbf{k}$ relative to q can be found in reference [23] with regard to optical transitions in semiconductors.

APPENDIX B: CALCULATION OF $\hat{\sigma}\hat{\sigma}^\dagger$

If we had not suppressed the $\mathbf{q}$ of $e^{i\mathbf{q}\cdot\mathbf{r}}$ in $\hat{H}'$ of Eq. (15b), we would have found that the $\hat{\sigma}$ and $\hat{\sigma}^\dagger$ of Eq. (26) would have to be chosen as

$$\hat{\sigma} = \frac{D_0}{\hbar\Omega}(\hat{\mathbf{p}} + \mathbf{p}_c)\cdot\boldsymbol{\lambda}^* e^{-i\mathbf{q}\cdot\mathbf{r}}, \quad \hat{\sigma}^\dagger = \frac{D_0}{\hbar\Omega}(\hat{\mathbf{p}} + \mathbf{p}_c)\cdot\boldsymbol{\lambda} e^{i\mathbf{q}\cdot\mathbf{r}}, \tag{B1}$$

such that to arrive at Eq. (25a),

$$\hat{\mathcal{H}} = \hat{\mathcal{H}}_0 - \hbar\Omega\hat{\sigma}\hat{\sigma}^\dagger. \tag{B2}$$

Consider the commutator $[\hat{\sigma}, \hat{\sigma}^\dagger]$ of the operators $\hat{\sigma}$ and $\hat{\sigma}^\dagger$, which are given in Eq. (26) for $\mathbf{q} \neq 0$,

$$[\hat{\sigma}, \hat{\sigma}^\dagger] = \left(\frac{D_0}{\hbar\Omega}\right)^2 [(\hat{\mathbf{p}} + \mathbf{p}_c)\cdot\boldsymbol{\lambda}^* e^{-i\mathbf{q}\cdot\mathbf{r}}, (\hat{\mathbf{p}} + \mathbf{p}_c)\cdot\boldsymbol{\lambda} e^{i\mathbf{q}\cdot\mathbf{r}}]. \tag{B3}$$

For the calculation, the commutator on the right-hand side of (B3) can be written as

$$\begin{aligned}[\hat{\sigma}, \hat{\sigma}^\dagger] = \left(\frac{D_0}{\hbar\Omega}\right)^2 \{&[(\hat{\mathbf{p}} + \mathbf{p}_c)\cdot\boldsymbol{\lambda}^* e^{-i\mathbf{q}\cdot\mathbf{r}}, (\hat{\mathbf{p}} + \mathbf{p}_c)\cdot\boldsymbol{\lambda}]e^{i\mathbf{q}\cdot\mathbf{r}} \\ &+ (\hat{\mathbf{p}} + \mathbf{p}_c)\cdot\boldsymbol{\lambda}[(\hat{\mathbf{p}} + \mathbf{p}_c)\cdot\boldsymbol{\lambda}^* e^{-i\mathbf{q}\cdot\mathbf{r}}, e^{i\mathbf{q}\cdot\mathbf{r}}] \\ &+ (\hat{\mathbf{p}} + \mathbf{p}_c)\cdot\boldsymbol{\lambda}^*[e^{-i\mathbf{q}\cdot\mathbf{r}}, (\hat{\mathbf{p}} + \mathbf{p}_c)\cdot\boldsymbol{\lambda}]e^{i\mathbf{q}\cdot\mathbf{r}} \\ &+ (\hat{\mathbf{p}} + \mathbf{p}_c)\cdot\boldsymbol{\lambda}[(\hat{\mathbf{p}} + \mathbf{p}_c)\cdot\boldsymbol{\lambda}^*, e^{i\mathbf{q}\cdot\mathbf{r}}]e^{-i\mathbf{q}\cdot\mathbf{r}}\}.\end{aligned} \tag{B4}$$

Furthermore, using the result that

$$\begin{aligned}[(\hat{\mathbf{p}} + \mathbf{p}_c)\cdot\boldsymbol{\lambda}^*, e^{i\mathbf{q}\cdot\mathbf{r}}] &= \hbar\mathbf{q}\cdot\boldsymbol{\lambda}^* e^{i\mathbf{q}\cdot\mathbf{r}}, \\ [e^{-i\mathbf{q}\cdot\mathbf{r}}, (\hat{\mathbf{p}} + \mathbf{p}_c)\cdot\boldsymbol{\lambda}] &= \hbar\mathbf{q}\cdot\boldsymbol{\lambda} e^{-i\mathbf{q}\cdot\mathbf{r}},\end{aligned} \tag{B5}$$

we can write the commutator in Eq. (B4) as

$$\begin{aligned}[\hat{\sigma}, \hat{\sigma}^\dagger] = \left(\frac{D_0}{\hbar\Omega}\right)^2 \{&(\hat{\mathbf{p}} + \mathbf{p}_c)\cdot\boldsymbol{\lambda}^*(\hbar\mathbf{q}\cdot\boldsymbol{\lambda}) \\ &+ (\hat{\mathbf{p}} + \mathbf{p}_c)\cdot\boldsymbol{\lambda}(\hbar\mathbf{q}\cdot\boldsymbol{\lambda}^*)\}.\end{aligned} \tag{B6}$$

Then we obtain

$$\begin{aligned}\hat{\sigma}\hat{\sigma}^\dagger = \hat{\sigma}^\dagger\hat{\sigma} + \left(\frac{D_0}{\hbar\Omega}\right)^2 \{&(\hat{\mathbf{p}} + \mathbf{p}_c)\cdot\boldsymbol{\lambda}^*(\hbar\mathbf{q}\cdot\boldsymbol{\lambda}) \\ &+ (\hat{\mathbf{p}} + \mathbf{p}_c)\cdot\boldsymbol{\lambda}(\hbar\mathbf{q}\cdot\boldsymbol{\lambda}^*)\}.\end{aligned} \tag{B7}$$

In the same way, we find

$$\hat{\sigma}^\dagger\hat{\sigma} = \left(\frac{D_0}{\hbar\Omega}\right)^2 (\hat{\mathbf{p}} + \mathbf{p}_c)\cdot\boldsymbol{\lambda} e^{i\mathbf{q}\cdot\mathbf{r}}(\hat{\mathbf{p}} + \mathbf{p}_c)\cdot\boldsymbol{\lambda}^* e^{-i\mathbf{q}\cdot\mathbf{r}}, \tag{B8}$$

and

$$(\hat{\mathbf{p}} + \mathbf{p}_c)\cdot\boldsymbol{\lambda}^* e^{-i\mathbf{q}\cdot\mathbf{r}} = -\hbar\mathbf{q}\cdot\boldsymbol{\lambda}^* e^{-i\mathbf{q}\cdot\mathbf{r}}, \tag{B9}$$

so that

$$\begin{aligned}\hat{\sigma}^\dagger\hat{\sigma} &= \left(\frac{D_0}{\hbar\Omega}\right)^2 (\hat{\mathbf{p}} + \mathbf{p}_c)\cdot\boldsymbol{\lambda}(-\hbar\mathbf{q}\cdot\boldsymbol{\lambda}^*) \\ &= -\left(\frac{D_0}{\hbar\Omega}\right)^2 \hbar\mathbf{q}\cdot\boldsymbol{\lambda}^*(\hat{\mathbf{p}} + \mathbf{p}_c)\cdot\boldsymbol{\lambda}.\end{aligned} \tag{B10}$$

Thus, finally, we find that when $\mathbf{q} \neq 0$,

$$\hat{\sigma}\hat{\sigma}^\dagger = \left(\frac{D_0}{\hbar\Omega}\right)^2 \hbar\mathbf{q}\cdot\boldsymbol{\lambda}(\hat{\mathbf{p}} + \mathbf{p}_c)\cdot\boldsymbol{\lambda}^* \tag{B11}$$

for use in Eq. (25a).

APPENDIX C: ANALYSIS OF THE MAGNITUDE OF D_0/Ω

Noting from Eqs. (14) and (15b), we see that both Ω and D_0 are naturally introduced and reoccur in a fundamentally key fashion in connection with the development physical observables throughout the paper. In particular, the magnitude of the ratio D_0/Ω becomes central to much of our analysis. Thus, from Eqs. (14) and (15b), we note that

$$\Omega = \omega\left(1 + \frac{\omega_p^2}{2\omega^2}\right), \tag{C1}$$

and, D_0 in magnitude,

$$|D_0| = \frac{|e|}{m_e c}\sqrt{\frac{2\pi\hbar c}{Vq}}, \tag{C2}$$

with $\omega = cq$. All other quantities are explicitly specified in Sec. II A.

Since Ω is already expressed in terms of the plasma frequency for an effective one-electron atom of metallic spacing; that is,

$$\omega_p = \sqrt{\frac{4\pi e^2}{m_e V}} \sim 10^{16} \text{ rad/s}, \tag{C3}$$

we can then similarly express $|D_0|$ of Eq. (C2) in terms of like variables as

$$|D_0| = \omega_p\left(\frac{\varepsilon_0}{\omega}\right)^{1/2}, \tag{C4}$$

where

$$\varepsilon_0 = \frac{\hbar}{2m_e}. \tag{C5}$$

It then follows from Eq. (C1) and (C4) that

$$\frac{|D_0|}{\Omega} = \Gamma\left(\frac{\omega_p}{\omega}\right)\sqrt{\frac{\varepsilon_0}{\omega}}, \tag{C6}$$

where

$$\Gamma(\omega_p/\omega) = \frac{\omega_p/\omega}{1 + \frac{1}{2}(\omega_p/\omega)^2}. \tag{C7}$$

We observe that $\Gamma(x)$ in Eq. (C7) is a universal function of x which peaks at $x = x_0 = \sqrt{2}$ with a value $\Gamma(x_0) = \Gamma_0 = 1/\sqrt{2}$. Thus, when ω is such that $\omega \gtrless \omega_0 = \omega_p/\sqrt{2}$, $\Gamma(\frac{\omega_p}{\omega})$ will fall away from the maximum at Γ_0. Then $|D_0|/\Omega$ in Eq. (C6) will vary as $\sqrt{\varepsilon_0/\omega}$.

We note that, in Eq. (C6), Γ is a dimensionless parameter whereas the square-root term has dimensions of length. For purposes of comparative analysis with respect to varying $\hbar\omega$, it is useful to normalize $|D_0|/\Omega$ to a dimensionless quantity. In this regard, in arriving at the approximation of Eq. (39), we expand $V_c(\mathbf{r} + i\hbar\hat{\boldsymbol{\alpha}}_0)$ to order $\hat{\boldsymbol{\alpha}}_0$, where from Eq. (27c) $\hat{\boldsymbol{\alpha}}_0 = (D_0/\hbar\Omega)(\boldsymbol{\lambda}^*\hat{a}^\dagger - \boldsymbol{\lambda}\hat{a})$, so that

$$V_c(\mathbf{r}+i\hbar\hat{\boldsymbol{\alpha}}_0) \simeq V_c(\mathbf{r})\big[1+i\hbar\hat{\boldsymbol{\alpha}}_0\cdot\nabla_{\mathbf{r}}V_c(\mathbf{r})/V_c(\mathbf{r})+O\big(\hat{\boldsymbol{\alpha}}_0^2\big)\big]. \tag{C8}$$

In estimating the magnitude of $\nabla_{\mathbf{r}}V_c(\mathbf{r})/V_c(\mathbf{r})$ with the inverse "average" lattice constant $1/a_L$, we can then express the magnitude of the renormalized constant as

$$\frac{|D_0|}{\Omega a_L} = \Gamma\left(\frac{\omega_p}{\omega}\right)\sqrt{\frac{a_0}{a_L}\frac{E_1}{\hbar\omega}} \simeq \sqrt{\Gamma^2\frac{0.15}{\hbar\omega(\mathrm{eV})}}; \tag{C9}$$

here, $a_0 = \frac{\hbar^2}{m_e e^2} \simeq 0.5$ Å, $a_L \simeq 5$ Å, and $E_1 = e^2/2a_L \simeq 1.5$ eV. $|D_0|/\Omega a_L$ of Eq. (C9) is much less than unity for values of $\hbar\omega$ ranging from the infrared to beyond the deep ultraviolet part of the electromagnetic spectrum.

APPENDIX D: BINOMIAL EXPANSION OF TWO NONCOMMUTING OPERATORS

We consider the binomial expansion of the function $(\hat{A}+\hat{B})^N/N!$ as presented in Eq. (48a) when $\hat{A}$ and $\hat{B}$ are noncommuting operators. To establish this consideration, it is appropriate to use the BCH theorem, written in Eq. (48b) as

$$e^{\lambda(\hat{A}+\hat{B})} = e^{\lambda\hat{A}}e^{\lambda\hat{B}}e^{-\frac{\lambda^2}{2}[\hat{A},\hat{B}]}, \tag{D1}$$

with $[\hat{A}[\hat{A},\hat{B}]] = [\hat{B}[\hat{A},\hat{B}]] = 0$.

We proceed by expanding the exponentials on the right-hand side of Eq. (D1) to obtain

$$e^{\lambda(\hat{A}+\hat{B})} = \sum_m \frac{1}{m!}\sum_{k=0}^{m}\binom{m}{k}\hat{A}^k\hat{B}^{m-k}\lambda^m e^{-\frac{\hat{C}}{2}}, \tag{D2}$$

where $\hat{C} = [\hat{A},\hat{B}]$. Then, in expanding $e^{\lambda(\hat{A}+\hat{B})}$ on the left-hand side, and $e^{-\frac{\lambda^2}{2}[\hat{A},\hat{B}]}$ (and combining λ^m to get λ^{m+2s}) on the right-hand side, we find that, in comparing with order λ^n, term by term,

$$\frac{(\hat{A}+\hat{B})^n}{n!} = \sum_{k=0,[(n-k)/2]}^{n}\frac{(-\hat{C}/2)^{\frac{n-k}{2}}}{k!\left(\frac{n-k}{2}\right)!}\sum_{r=0}^{k}\binom{k}{r}\hat{A}^r\hat{B}^{k-r}. \tag{D3}$$

Here, $[(n-k)/2]$ means that $(n-k)/2$ must be an integer. For $\hat{A}$, $\hat{B}$ as noted in Eqs. (44a) and (44b), Eq. (D3) reduces to Eq. (49).

APPENDIX E: EVALUATION OF $\langle m|\hat{a}^r(\hat{a}^\dagger)^{k-r}|m'\rangle$

To complete the evaluation of Eq. (49), and thus move ahead to the evaluation of $\langle m'|\hat{Q}V_c(\mathbf{r})|m\rangle$ in Eq. (50a), we find it necessary to establish the matrix element of the product operator $\hat{a}^r(\hat{a}^\dagger)^{k-r}$ with respects to the states (m, m'). We accomplish this through the decomposition

$$\langle m|\hat{a}^r(\hat{a}^\dagger)^{k-r}|m'\rangle = \sum_{m''}\langle m|\hat{a}^r|m''\rangle\langle m''|(\hat{a}^\dagger)^{k-r}|m'\rangle. \tag{E1}$$

It is straightforward to show that

$$\langle m|\hat{a}^r|m''\rangle = \sqrt{\frac{m''!}{(m''-r)!}}\,\delta_{m,m''-r}, \tag{E2a}$$

$$\langle m''|(\hat{a}^\dagger)^{k-r}|m'\rangle = \sqrt{\frac{(m'+k-r)!}{m'!}}\,\delta_{m'',m'+k-r}. \tag{E2b}$$

It then follows using Eqs. (E2a) and (E2b) that Eq. (E1) becomes

$$\begin{aligned}\langle m|\hat{a}^r(\hat{a}^\dagger)^{k-r}|m'\rangle &= \sum_{m''}\sqrt{\frac{m''!}{(m''-r)!}}\\ &\quad\times\sqrt{\frac{(m'+k-r)!}{m'!}}\,\delta_{m,m''-r}\delta_{m'',m'+k-r}\\ &= \sqrt{\frac{(m+r)!(m'+k-r)!}{m!m'!}}.\end{aligned} \tag{E3}$$

From the selection rules of Eqs. (E2a) and (E2b), it is clear that eliminating m'' we get $m+r = m'+k-r$. Thus, we find

$$\langle m|\hat{a}^r(\hat{a}^\dagger)^{k-r}|m'\rangle = \frac{(m+r)!}{\sqrt{m!m'!}}. \tag{E4}$$

APPENDIX F: DENSITY MATRIX FORMULATION OF CURRENT

In developing the current from the density matrix [24] for our two Hamiltonians, we have

$$\begin{aligned}\mathbf{j}(t) &= e\sum_{n,\mathbf{k},m}\sum_{n',\mathbf{k}',m'}\hat{\varrho}_{n\mathbf{k}m,n'\mathbf{k}'m'}(t)\hat{\mathbf{v}}_{n'\mathbf{k}'m',n\mathbf{k}m}(t)\\ &= eTr(\hat{\varrho}\hat{\mathbf{v}}),\end{aligned} \tag{F1}$$

where the matrix elements of $\hat{\mathbf{v}}$ are given by Eqs. (62a) and (62b), depending upon which case we are developing. The matrix elements of $\hat{\varrho}$, the density-matrix operator, are governed by the Liouville equation

$$i\hbar\frac{\partial\hat{\varrho}}{\partial t} = [\hat{H},\hat{\varrho}]. \tag{F2}$$

Using the form for $\hat{H}$ given for the two canonical Hamiltonians in Sec. II, i.e., $\hat{H} = \hat{H}_0 + \hat{H}'$, with identical $\hat{H}_0$ but differing $\hat{H}'$, we use of the instantaneous Bloch eigenstates and harmonic-oscillator eigenstates as basis states to further develop the Liouville equation evaluation. As such, we find

$$i\hbar\left(\frac{\partial\hat{\varrho}}{\partial t}\right)_{n'\mathbf{k}'m',n\mathbf{k}m} = [\hat{H}_0,\hat{\varrho}]_{n'\mathbf{k}'m',n\mathbf{k}m} + [\hat{H}',\hat{\varrho}]_{n'\mathbf{k}'m',n\mathbf{k}m},$$

which, using the properties of $\psi_{n\mathbf{K}}$ and $|m\rangle$, evolves to

$$\begin{aligned}&i\hbar\left(\frac{\partial\hat{\varrho}}{\partial t}\right)_{n'\mathbf{k}'m',n\mathbf{k}m}\\ &\quad= [\varepsilon_{n'}(\mathbf{k}')-\varepsilon_n(\mathbf{k})+E_{m'}-E_m]\hat{\varrho}_{n'\mathbf{k}'m',n\mathbf{k}m}\\ &\qquad+\sum_{n'',\mathbf{k}'',m''}\big[\hat{H}'_{n'\mathbf{k}'m',n''\mathbf{k}''m''}\hat{\varrho}_{n''\mathbf{k}''m'',n\mathbf{k}m}\\ &\qquad-\hat{\varrho}_{n'\mathbf{k}'m',n''\mathbf{k}''m''}\hat{H}'_{n''\mathbf{k}''m'',n\mathbf{k}m}\big].\end{aligned} \tag{F3}$$

The last term in Eq. (F3) results from the insertion of the "double prime" complete set of functions.

Taking into account that the instantaneous eigenstates are time dependent and they satisfy

$$i\hbar\frac{\partial\psi_{n\mathbf{K}}(\mathbf{r},t)}{\partial t} = \mathbf{F}\cdot\sum_{n''}\mathbf{R}_{n''n}[\mathbf{k}(t)]\psi_{n''\mathbf{K}}(\mathbf{r},t),$$

it follows that

$$i\hbar\left(\frac{\partial\hat{\varrho}}{\partial t}\right)_{n'\mathbf{k}'m',n\mathbf{k}m} = i\hbar\frac{\partial}{\partial t}\hat{\varrho}_{n'\mathbf{k}'m',n\mathbf{k}m} + \mathbf{F}\cdot\sum_{n''}[\mathbf{R}_{n''n'}(\mathbf{k}')\hat{\varrho}_{n''\mathbf{k}'m',n\mathbf{k}m} - \mathbf{R}_{n''n}(\mathbf{k})\hat{\varrho}_{n'\mathbf{k}'m',n''\mathbf{k}m}].$$

Putting this into (F3), we get

$$i\hbar\frac{\partial}{\partial t}\hat{\varrho}_{n'\mathbf{k}'m',n\mathbf{k}m} = [\mathcal{E}_{n'}(\mathbf{k}') - \mathcal{E}_n(\mathbf{k}) + E_{m'} - E_m] \times\hat{\varrho}_{n'\mathbf{k}'m',n\mathbf{k}m} + \hat{H}''_{n'\mathbf{k}'m',n\mathbf{k}m}[\hat{\varrho}_{n\mathbf{k}m} - \hat{\varrho}_{n'\mathbf{k}'m'}] + \sum_{n'',\mathbf{k}'',m''}{}' \left[\hat{H}''_{n'\mathbf{k}'m',n''\mathbf{k}''m''}\hat{\varrho}_{n''\mathbf{k}''m'',n\mathbf{k}m} - \hat{\varrho}_{n'\mathbf{k}'m',n''\mathbf{k}''m''}\hat{H}''_{n''\mathbf{k}''m'',n\mathbf{k}m}\right]. \quad \text{(F4)}$$

Here, $\hat{\varrho}_{n\mathbf{k}m} = \hat{\varrho}_{n\mathbf{k}m,n\mathbf{k}m}$, $\mathcal{E}_n(\mathbf{k}) = \varepsilon_n(\mathbf{k}) + \hat{H}''_{n\mathbf{k}m,n\mathbf{k}m}$, and

$$\hat{H}''_{n'\mathbf{k}'m',n''\mathbf{k}''m''} = \hat{H}'_{n'\mathbf{k}'m',n''\mathbf{k}''m''} - \mathbf{F}(t)\cdot\mathbf{R}_{n'n''}(\mathbf{k}')\delta_{\mathbf{k}',\mathbf{k}''}\delta_{m',m''}.$$

In Eq. (F4), the off-diagonal elements and diagonal elements of $\hat{\varrho}$ have been separated; the $\sum'$ indicates only off-diagonal elements of $\hat{\varrho}$. In Eq. (F4), we now drop the term $\sum'$ which contains higher-order terms in $\hat{H}''$. Then, we can solve the remaining equation for $\hat{\varrho}_{n'\mathbf{k}'m',n\mathbf{k}m}$, the off-diagonal matrix elements, as

$$\hat{\varrho}_{n'\mathbf{k}'m',n\mathbf{k}m}(t) = \frac{1}{i\hbar}\int_{t_0}^{t} dt'\hat{H}''_{n'\mathbf{k}'m',n\mathbf{k}m}(t')[\hat{\varrho}_{n\mathbf{k}m}(t') - \hat{\varrho}_{n'\mathbf{k}'m'}(t')] \times e^{\frac{i}{\hbar}\int_t^{t'}[\mathcal{E}_{n'}(\mathbf{k}')-\mathcal{E}_n(\mathbf{k})+E_{m'}-E_m]d\tau}, \quad \text{(F5)}$$

with $\hat{\varrho}_{n'\mathbf{k}'m',n\mathbf{k}m}(t_0) = 0$, $n'\mathbf{k}'m' \neq n\mathbf{k}m$ being an approximate expression for the off-diagonal matrix elements.

In Eq. (F4), if we consider $n'\mathbf{k}'m' = n\mathbf{k}m$, then we can write

$$i\hbar\frac{\partial}{\partial t}\hat{\varrho}_{n\mathbf{k}m} = \sum_{n'',\mathbf{k}'',m''}{}' \left[\hat{H}''_{n\mathbf{k}m,n''\mathbf{k}''m''}\hat{\varrho}_{n''\mathbf{k}''m'',n\mathbf{k}m} + \hat{\varrho}_{n\mathbf{k}m,n''\mathbf{k}''m''}\hat{H}''_{n''\mathbf{k}''m'',n\mathbf{k}m}\right] = \sum_{n',\mathbf{k}',m'}{}' \left[(\hat{H}'')^*_{n'\mathbf{k}'m',n\mathbf{k}m}\hat{\varrho}_{n'\mathbf{k}'m',n\mathbf{k}m} + \hat{H}''_{n\mathbf{k}m,n'\mathbf{k}'m'}\hat{\varrho}^*_{n'\mathbf{k}'m',n\mathbf{k}m}\right]. \quad \text{(F6)}$$

Then, in eliminating the off-diagonal matrix elements in Eq. (F6) using (F5), we obtain a closed equation for the diagonal matrix elements

$$\frac{\partial}{\partial t}\hat{\varrho}_{n\mathbf{k}m}(t) = \frac{2}{\hbar^2}\mathrm{Re}\sum_{n',\mathbf{k}',m'}{}' \left[\hat{H}''_{n\mathbf{k}m,n'\mathbf{k}'m'}(t) \times\int_{t_0}^{t} dt'\hat{H}''_{n'\mathbf{k}'m',n\mathbf{k}m}(t')e^{\frac{i}{\hbar}\int_t^{t'}[\mathcal{E}_{n'}(\mathbf{k}')-\mathcal{E}_n(\mathbf{k})+E_{m'}-E_m]d\tau} \times[\hat{\varrho}_{n'\mathbf{k}'m'}(t') - \hat{\varrho}_{n\mathbf{k}m}(t')]\right]. \quad \text{(F7)}$$

Thus, in (F5) and (F7), we have developed approximate expressions for the off-diagonal and diagonal density-matrix elements for our problem at hand. In evaluating the matrix elements of the velocity and determining the diagonal and off-diagonal components, we can then apply the appropriate diagonal or off-diagonal density matrix elements from Eqs. (F5) and (F7) to evaluate the current from Eq. (F1), that is

$$\mathbf{j}(t) = e\left[\sum_{n,\mathbf{k},m}\hat{\varrho}_{n\mathbf{k}m}(t)\hat{\mathbf{v}}_{n\mathbf{k}m,n\mathbf{k}m}(t) + \sum_{n,\mathbf{k},m}\sum_{\substack{n',\mathbf{k}',m'\\ \neq n,k,m}}\hat{\varrho}_{n\mathbf{k}m,n'\mathbf{k}'m'}(t)\hat{\mathbf{v}}_{n'\mathbf{k}'m',n\mathbf{k}m}(t)\right]. \quad \text{(F8)}$$

APPENDIX G: MULTIBAND APPROACH TO EQ. (65) USING WIGNER-WEISSKOPF APPROXIMATION

In seeking an approximate solution to Eqs. (65) or (66), we have looked for a first-order solution in the form of Eq. (67a) which results in Eq. (67b). We adopted this approach since the solutions to Eqs. (65) and (66) are not tractable. But we explore here an alternative methodology [25] for obtaining an improved solution to Eq. (65). In this regard, we express Eq. (65) in the form

$$i\hbar\dot{C}_{n\mathbf{k}}(m,t) = -\sum_{n',\mathbf{k}',m'}B_{n\mathbf{k}m,n'\mathbf{k}'m'}(t)C_{n'\mathbf{k}'}(m',t), \quad \text{(G1)}$$

where

$$B_{n\mathbf{k}m,n'\mathbf{k}'m'}(t) = \mathbf{F}(t)\cdot\mathbf{R}_{nn'}(\mathbf{k}')e^{\frac{i}{\hbar}\int_{t_0}^{t}[\varepsilon_n(\mathbf{k})-\varepsilon_{n'}(\mathbf{k}')]d\tau}\delta_{\mathbf{k},\mathbf{k}'}\delta_{m,m'} - \hat{\mathcal{H}}'_{n\mathbf{k},n'\mathbf{k}'}(m,m',t)e^{\frac{i}{\hbar}\int_{t_0}^{t}[\varepsilon_n(\mathbf{k})-\varepsilon_{n'}(\mathbf{k}')+E_m-E_{m'}]d\tau}. \quad \text{(G2)}$$

Now, approximating $C_{n'\mathbf{k}'}(m',t)$ on the right-hand side of Eq. (G1) as

$$i\hbar\dot{C}_{n'\mathbf{k}'}(m',t) = -B_{n'\mathbf{k}'m',n\mathbf{k}m}(t)C_{n\mathbf{k}}(m,t), \quad \text{(G3)}$$

where $B_{n'\mathbf{k}'m',n\mathbf{k}m}(t) = B^*_{n\mathbf{k}m,n'\mathbf{k}'m'}(t)$, it follows that Eq. (G1) becomes

$$\dot{C}_{n\mathbf{k}}(m,t) = -\frac{1}{\hbar^2}\sum_{n',\mathbf{k}',m'}B_{n\mathbf{k}m,n'\mathbf{k}'m'}(t) \times\int_{t_0}^{t} dt'B^*_{n\mathbf{k}m,n'\mathbf{k}'m'}(t')C_{n\mathbf{k}}(m,t'). \quad \text{(G4)}$$

Equations (G1) and (G3) are noted as the Wigner-Weisskopf [25] approximation (WWA); in essence, the WWA couples the state $n\mathbf{k}m$ to all states $n'\mathbf{k}'m'$ while including only the direct reflective feedback from each $n'\mathbf{k}'m'$ state back to the state of interest. This approximation guarantees conservation of probability, since it can be shown that [26]

$$|C_{n\mathbf{k}}(m,t)|^2 + \sum_{n'\mathbf{k}'m'\neq n\mathbf{k}m}|C_{n'\mathbf{k}'}(m',t)|^2 = 1, \quad \text{(G5)}$$

when $C_{n\mathbf{k}}(m,t=t_0) = 1$ and $C_{n'\mathbf{k}'}(m',t=t_0) = 0$, $n'\mathbf{k}'m' \neq n\mathbf{k}m$. Finally, we see that by using the WWA, the original set of coupled equations for $C_{n\mathbf{k}}(m,t)$ has been reduced to $n\mathbf{k}m$ uncoupled integro-differential equations. In pursuing the analysis, Eq. (G4) might be a fruitful approach for calculating more accurate Bloch current in Eq. (68).

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