Computer Physics Communications ▮ (▮▮▮▮) ▮▮▮–▮▮▮

Contents lists available at ScienceDirect

Computer Physics Communications

journal homepage: www.elsevier.com/locate/cpc

Adiabatic corrections for velocity-gauge simulations of electron dynamics in periodic potentials

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ARTICLE INFO

Article history:
Received 28 February 2017
Received in revised form 19 April 2017
Accepted 23 April 2017
Available online xxxx

Keywords:
Electron dynamics in solids
Strong laser fields
Quantum mechanical
Polarization response
Velocity gauge

ABSTRACT

We show how to significantly reduce the number of energy bands required to model the interaction of light with crystalline solids in the velocity gauge. We achieve this by deriving analytical corrections to the electric current density. These corrections depend only on band energies, the matrix elements of the momentum operator, and the macroscopic vector potential. Thus, the corrections can be evaluated independently from modeling the interaction with light. In addition to improving the convergence of velocity-gauge calculations, our analytical approach overcomes the long-standing problem of divergences in expressions for linear and nonlinear susceptibilities.

1. Introduction

The velocity and length gauges are two most frequent choices for a theoretical description of the interaction between electromagnetic radiation and matter. Even though, in principle, all physical observables must be gauge-independent, the gauge choice is very important for numerical simulations because approximations and discretization errors violate the gauge invariance in its strict sense [1]. In atomic and molecular physics, where quantum systems are finite, both gauges are easy to implement. The situation is different for modeling the interaction of light with bulk crystals, where an infinite periodic lattice potential and the degenerate energy states present difficulties for length-gauge implementations [2,3]. These difficulties also pertain to the closely related approach of modeling quantum dynamics in the basis of Houston states [4], also known as accelerated Bloch states [5]. The fundamental problem here is that these methods require differentiation with respect to the crystal momentum, $\mathbf{k}$, while numerically evaluated Bloch states are, in general, not smooth functions of $\mathbf{k}$ [6,7]. The problem becomes particularly severe for modeling strong-field phenomena, where electrons traverse a significant part of the Brillouin zone during a laser cycle [8–11]. Solutions to these problems are known [3,12], but they either do not ensure the periodicity with respect to $\mathbf{k}$ [13] or require the evaluation of so-called covariant derivatives [3,14,15]. Evaluating covariant derivatives with respect to the crystal momentum at each propagation step slows down computations. In contrast, the velocity gauge does not require differentiation with respect to the crystal momentum, which often makes it advantageous for modeling light-driven electron dynamics [16–21]. However, performing velocity-gauge simulations in a basis of Bloch states has a serious drawback: for convergence, they usually require many more energy bands than length-gauge or Houston-basis simulations [22]. In this paper, we propose a solution to this problem. We do so by deriving analytical corrections to the polarization response evaluated with a relatively small number of bands (Section 3). Although we derive our corrections in the limit of a weak external field, we show in Section 4 that they work surprisingly well even for strong optical fields.

2. Velocity-gauge description of light–solid interaction

Let $|n\mathbf{k}\rangle$ denote a Bloch state with band index n and crystal momentum $\mathbf{k}$. The Bloch states are eigenstates of an unperturbed Hamiltonian $\hat{H}^{(0)}$. That is, $\hat{H}^{(0)}|n\mathbf{k}\rangle = \epsilon_n(\mathbf{k})|n\mathbf{k}\rangle$, where $\epsilon_n(\mathbf{k})$ is energy. Even though $\hat{H}^{(0)}$ is a single-particle operator, it may represent an outcome of self-consistent many-body calculations. We describe the interaction with a classical electromagnetic field by adding an interaction operator, $\hat{H}_{\text{int}}$, to the Hamiltonian. Unitary transformations allow one to write $\hat{H}_{\text{int}}$ in different forms. One of them is $\hat{H}_{\text{int}} = e(\mathbf{A}\cdot\hat{\mathbf{p}} + \hat{\mathbf{p}}\cdot\mathbf{A})/(2m_0) + e^2A^2/m_0$. Here, $\mathbf{A}$ is the vector potential of the electromagnetic field, $\hat{\mathbf{p}}$ is the momentum operator, $e > 0$ is the elementary charge, and m_0 is the electron mass. In SI units, the relation between $\mathbf{A}$ and the electric field is $\mathbf{E} = -\partial\mathbf{A}/\partial t$. In the following, we assume the

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http://dx.doi.org/10.1016/j.cpc.2017.04.010
0010-4655/

dipole approximation, where the dependence of $\mathbf{A}$ on coordinate, $\mathbf{r}$, is neglected. The term e^2A^2/m_0 can be eliminated by the unitary transformation $|\psi\rangle \rightarrow \exp[-ie^2/(2\hbar m_0)\int_{-\infty}^{t} A^2(t')\mathrm{d}t']\,|\psi\rangle$. The result is the velocity-gauge form of the Hamiltonian:

$$\hat{H}(t) = \hat{H}^{(0)} + \hat{H}_{\mathrm{int}} = \hat{H}^{(0)} + \frac{e}{m_0}\mathbf{A}(t)\cdot\hat{\mathbf{p}}. \tag{1}$$

In this equation, the external field preserves the spatial periodicity of the lattice potential. Therefore, the Bloch theorem applies not only to $\hat{H}^{(0)}$, but also to $\hat{H}(t)$. Consequently, transitions induced by the external field formally preserve the crystal momentum, which is one of the most important reasons why velocity-gauge simulations are so attractive for numerical calculations.

However, the convenience of preserving the crystal momentum comes at a price. This becomes apparent if we consider the interaction of a dielectric with a constant electric field ($\mathbf{E} = \mathrm{const}$) that is sufficiently weak to neglect interband tunneling. Even though, in the absence of free charge carriers, all physical observables are expected to take constant values, the velocity-gauge wave functions have a nontrivial dependence on time because the vector potential, $\mathbf{A}(t) = -\int_{-\infty}^{t}\mathbf{E}(t')\mathrm{d}t'$, makes the Hamiltonian time-dependent. In this case, numerical approximations, such as truncating the basis, lead to a spurious divergence of the polarization response in the low-frequency limit [2,23–26]. This is one of the underlying reasons why velocity-gauge calculations require many more bands than length-gauge or Houston-basis ones.

Another, closely related, problem with the velocity gauge can be understood by considering a particular Bloch state exposed to a weak long half-cycle of the electric field. Because the field performs only a half of the oscillation, the vector potential has a nonzero value at the end of the pulse. In the adiabatic limit, the pulse transforms an initial state $|n\mathbf{k}_0\rangle$ into another Bloch state, $|n\mathbf{k}\rangle$, provided that the nth band is nondegenerate along the reciprocal-space path prescribed by the acceleration theorem: $\mathbf{k}(t) = \mathbf{k}_0 + e\hbar^{-1}\mathbf{A}(t)$. This adiabatic intraband motion is guaranteed in length-gauge and Houston-basis calculations, making these approaches a good choice when only ad hoc transition matrix elements are available [27,28]. In contrast, the correct adiabatic behavior is not explicitly encoded in the velocity-gauge equations of motion—it is implicit in the parameters of the model, demanding accurate energies and matrix elements.

The only matrix elements required in the velocity-gauge simulations are those of the momentum operator:

$$\mathbf{p}_{nm}(\mathbf{k}) = \frac{1}{\Omega}\int_{\mathrm{cell}} \mathrm{d}^3(\mathbf{r})\,\psi^*_{n\mathbf{k}}(\mathbf{r})\hat{\mathbf{p}}\psi_{m\mathbf{k}}(\mathbf{r}), \tag{2}$$

where the integration is performed over a unit cell, Ω is the cell volume, and $\psi_{n\mathbf{k}}(\mathbf{r}) = \langle\mathbf{r}|n\mathbf{k}\rangle$ are Bloch functions in the coordinate representation. Indeed, let us search for time-dependent wave functions using the following ansatz:

$$|\psi_{\mathbf{k}}(t)\rangle = \sum_m \alpha_{m\mathbf{k}}(t)\,|m\mathbf{k}\rangle\,. \tag{3}$$

Inserting this ansatz into the time-dependent Schrödinger equation (TDSE), $\mathrm{i}\hbar\partial_t\,|\psi\rangle = \hat{H}(t)\,|\psi\rangle$, and assuming that $\langle m\mathbf{k}'|n\mathbf{k}\rangle = \delta_{mn}\delta(\mathbf{k}-\mathbf{k}')$, we obtain a system of coupled differential equations for the probability amplitudes:

$$\mathrm{i}\hbar\frac{\partial\,\alpha_{q\mathbf{k}}}{\partial t} = \epsilon_q(\mathbf{k})\alpha_{q\mathbf{k}} + \frac{e}{m_0}\mathbf{A}(t)\cdot\sum_m \mathbf{p}_{qm}(\mathbf{k})\alpha_{m\mathbf{k}}. \tag{4}$$

In these equations, the medium properties are fully defined by $\epsilon_q(\mathbf{k})$ and $\mathbf{p}_{qm}(\mathbf{k})$. Once the probability amplitudes are evaluated, the electric current density can be obtained according to

$$\mathbf{J}(t) = \sum_{n\in\mathrm{occ}}\int_{\mathrm{BZ}}\frac{\mathrm{d}^3\mathbf{k}}{(2\pi)^3}f_n(\mathbf{k})\mathbf{j}_{n\mathbf{k}}(t). \tag{5}$$

Here, $f_n(\mathbf{k})$ is the Fermi factor of band n in the initial (stationary) state. The summation in Eq. (5) is carried out over all the bands that contain electrons in the unperturbed state (these would be valence bands for a dielectric), the integration is performed over the first Brillouin zone (BZ), and the contribution from crystal momentum $\mathbf{k}$ in initial band n is given by

$$\mathbf{j}_{n\mathbf{k}}(t) = -\frac{e}{m_0}\left(e\mathbf{A}(t) + \mathrm{Re}\left[\sum_{ij}\left(\alpha^{(n)}_{i\mathbf{k}}(t)\right)^*\alpha^{(n)}_{j\mathbf{k}}(t)\mathbf{p}_{ij}(\mathbf{k})\right]\right). \tag{6}$$

In this equation, we have added a superscript to the probability amplitudes in order to denote the initial band: $\alpha^{(n)}_{i\mathbf{k}}(t_{\mathrm{min}}) = \delta_{in}$. That is, in each initial state, only one band is occupied; by adding the contributions from all the initial states, as prescribed by Eq. (5), we model a solid with occupied valence bands. Once $\mathbf{J}(t)$ is evaluated, the polarization response, which is the key quantity in linear and nonlinear optics, is readily given by [29,30]

$$\mathbf{P}(t) = \int_{-\infty}^{t}\mathrm{d}t'\,\mathbf{J}(t'). \tag{7}$$

3. Velocity-gauge corrections

In the previous section, we argued that velocity-gauge simulations tend to violate adiabaticity where adiabaticity is expected, which is why they demand high-quality band energies and matrix elements. In this section, we derive analytical corrections that alleviate this issue in the case where a finite number of energy bands is the main source of discretization errors. In the next section, we will demonstrate that our corrections work well even if the optical response is far from being adiabatic (that is, the polarization $\mathbf{P}(t)$ is not a function of the electric field at time t). In spite of that, we derive the corrections by considering an adiabatic limit. It is worth clarifying this point before we start the derivation. If velocity-gauge simulations require N_{VG} bands to produce physically meaningful results, while length-gauge simulations require only $N_{\mathrm{LG}} < N_{\mathrm{VG}}$ bands, then the bands that are unnecessary in the length gauge serve a rather numerical than physical purpose. They are required in the velocity gauge to ensure that the basis set is reasonably complete. After the interaction with an external field, the occupation of these highly excited states is negligibly small. Even if the polarization response due to "physically relevant" bands is very nontrivial, the contributions due to very high conduction bands (and very low valence ones) are expected to be, in some sense, "simple". We argue that this simplicity consists in the adiabaticity with respect to the vector potential: the difference between the exact current density, $\mathbf{J}(t)$, and that evaluated with a finite number of bands, $\mathbf{J}_N(t)$ is, approximately, a function of the vector potential, $\mathbf{A}$, at time t. In this case, one may use any function $\mathbf{A}(t)$ that is particularly well suited for analytical calculations. We chose

$$\mathbf{A}(t) = \mathrm{Re}\left[\mathbf{a}\mathrm{e}^{\gamma t - \mathrm{i}\omega_0 t}\right], \tag{8}$$

where $\mathbf{a}$ is a constant vector (it is complex-valued unless $\mathbf{A}$ is linearly polarized), ω_0 is a carrier-wave frequency, and $\gamma > 0$ is a small parameter that controls how slowly the field is turned on. The next step will be the evaluation of the current density using time-dependent perturbation theory, where we only consider a time interval where the external field may be viewed as a perturbation. Once this step is accomplished, we will take the adiabatic limit: $\omega_0 \rightarrow 0$. Here comes our key idea. When the amplitude of the vector potential, $|\mathbf{a}|$, is fixed and its frequency is decreased, then the electric field decreases as well; consequently, we expect $\lim_{\gamma\rightarrow 0+}\lim_{\omega_0\rightarrow 0}\mathbf{J}(t) \equiv 0$ for a dielectric and, more generally,

$$\lim_{\gamma\rightarrow 0+}\lim_{\omega_0\rightarrow 0}\mathbf{J}(t) \equiv \mathbf{J}_{\mathrm{ad}}\big(\mathbf{A}(t)\big) \tag{9}$$

Please cite this article in press as: V.S. Yakovlev, M.S. Wismer, Adiabatic corrections for velocity-gauge simulations of electron dynamics in periodic potentials, Computer Physics Communications (2017), http://dx.doi.org/10.1016/j.cpc.2017.04.010.