Gauge Invariant Formulation of the Semiconductor Bloch Equations

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We derive gauge invariant semiconductor Bloch equations (GI-SBEs) that contain only gauge invariant band structure, shift vectors, and triple phase products. The validity and utility of the GI-SBEs is demonstrated in intense laser driven solids with broken inversion symmetry and nontrivial topology. The GI-SBEs present a useful platform for modeling and interpreting light-matter interactions in solids, in which the gauge freedom of the Bloch basis functions obscures physics and creates numerical obstacles.

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High harmonic generation (HHG) in solids was first observed in a bulk semiconductor [1], and more recently has been explored in a wider variety of materials such as dielectrics [2,3], two-dimensional semiconductors [4,5], and nanostructures [6,7]. Its discovery has laid the foundation of attosecond science in condensed matter [8–12], and has enabled optical reconstruction of band structural properties such as energy bands, [13], transition dipole moments (TDMs) [14], and Berry curvature [15–17].

Several theoretical approaches have been developed to model HHG in solids, ranging from semiclassical analysis [18–20] to various quantum mechanical simulations—the latter including time-dependent density-functional theory [21,22], as well as the semiconductor Bloch equations (SBEs) [23–28] in length and velocity gauge, and Wannier basis analysis [29].

The SBEs in length gauge and within the one-body approximation have been used extensively and successfully to model HHG in solids [13,18–20,27,29–40]. First, they allow the laser induced currents to be interpreted within the Bloch band picture, providing for an intuitive decomposition into intraband and interband contributions [27,41,42]. Second, phenomenological relaxation mechanisms due to many-body effects are commonly added in the length gauge; in other approaches, such as in the velocity gauge, they can lead to nonphysical results [43].

Bloch functions, the eigenfunctions of a periodic solid, exhibit a phase freedom and we refer to a specific choice of phases as a Bloch gauge. This constitutes a unitary gauge symmetry that is analogous to, but distinct from, electromagnetic gauge symmetry [44]. Bloch gauge freedom causes difficulties in crystals with broken inversion or time-reversal symmetry, as the SBEs involve Berry connections and complex-valued TDMs, both of which are gauge dependent [33,34,36]. Separating their individual contributions to experimental features is not possible, since they are not uniquely defined. Further, numerical structural calculations determine Bloch eigenfunctions up to an arbitrary phase, which poses the problem of finding a smooth Bloch gauge for numerical analysis of the SBEs [27,37,38,45].

Numerical approaches based on the parallel transport gauge transformation [46] have been demonstrated successfully [37], but the implementation can be quite challenging, especially in systems with degeneracies, or for driving lasers with time-dependent polarization [38]. This method is also inadequate for applications requiring gauge fixing of all components of the Berry connections, such as tomographic reconstruction of the TDM in multiple dimensions. Further, materials with nontrivial topology do not admit a globally smooth Bloch gauge [47,48], and thus require a different approach to overcome the numerical obstacles resulting from gauge ambiguity.

To reap the benefits of length gauge SBEs without its shortcomings, SBEs need to be found that contain only Bloch gauge invariant quantities. This would be analogous to Maxwell’s equations which can be formulated in terms of physically measurable, gauge invariant electric and magnetic fields (length gauge), or in terms of gauge-dependent potentials (velocity gauge).

In this paper we formulate the gauge invariant SBEs (GI-SBEs). This represents progress on four levels. (i) On a fundamental level, the GI-SBEs contain only gauge invariant quantities, in analogy to Maxwell’s equations for electric and magnetic fields; (ii) On a technical level, gauge invariance eliminates the need for numerical gauge smoothing; (iii) On a physics related level, we find that in the GI-SBEs broken inversion symmetry and topological effects manifest themselves in gauge invariant shift vectors and triple phase products. In particular, the shift vector has been studied previously for perturbative nonlinear optics [49–52], and more recently within the semiclassical approximation for HHG [40]. The GI-SBEs complement and generalize previous work by providing a theoretical and quantitative framework that is not restricted to any specific optical regime; (iv) On an
The SBEs for the density matrix are \[ \frac{\partial}{\partial t} \rho_{nm}(\mathbf{k}, t) = -e \mathbf{F}(t) \cdot \mathbf{D}, \rho(t) \] where the energy difference between bands, \[ E_{nm} = E_n - E_m \] is the energy difference between bands. The SBEs for the off-diagonal components of the coherence matrix are

\[ \frac{\partial}{\partial t} \mathbf{q}_{nm}(\mathbf{k}, t) = -e \mathbf{F}(t) \cdot \nabla_{\mathbf{k}} \mathbf{q}_{nm} - 2e \mathbf{F}(t) \sum_{l} [d_{nl}] \text{Im} |q_{lm}|. \]
gauge invariant quantities. The intraband current involves populations and band velocities, both of which are gauge invariant. In contrast, interband HHG involves the gauge covariant TDM and density matrix. To evaluate the interband current within the gauge invariant formalism, we use Eq. (4) and write each vector component as

$$j_{ct,i}(t) = e \sum_{n\neq m} \int d\mathbf{k} e^{\pi} \langle \mathbf{d}_{nm,i}(\mathbf{k}) | e^{\pi} \mathbf{q}_{mn}(\mathbf{k}, t) \rangle,$$

(9)

where $d_{nm,i} = e_i \cdot d_{nm}$ is the TDM component along emission direction $e_i$, and $\phi_{nm,i} = \operatorname{arg}[d_{nm,i}]$. The phase difference $\theta_{nm,i} \equiv \phi_{nm,i} - \phi_{nm}$ is gauge invariant, since all vector components of the TDM transform equivalently under Bloch phase rotations. Further, $\theta_{nm,i} = 0$ for the direction parallel to the laser polarization, and the off-diagonal components of $\theta$ represent the material polarization along $n$, since $n \cdot \mathbf{P}(t) = e \sum_{n \neq m} \int d\mathbf{k} \mathbf{q}_{nm}(\mathbf{k}, t) | d_{nm}(\mathbf{k}) |$.

To demonstrate the validity and utility of the GI-SBEs, we simulate HHG in select systems and compare the results to the more familiar GC-SBEs. It is essential to consider systems with broken inversion or time-reversal symmetry, as the shift vector vanishes if both symmetries are present [40].

First we simulate HHG in bulk ZnO. The band structure is obtained from an empirical tight-binding model [57]; for simplicity we include only the highest valence ($v$) and the two lowest conduction bands ($c_1$, $c_2$), and model HHG along the $\Gamma L$ symmetry axis of the BZ, as illustrated in the SM [53]. The matrix elements required for the GC-SBEs and GI-SBEs are obtained numerically. The gauge-dependent Berry connections and TDM phases are calculated using the twisted parallel transport gauge smoothing algorithm [37,46], and the results for these quantities are shown in the SM [53]. The shift vectors are determined via a generalized Wilson loop algorithm [58], while the TDM magnitudes and triple product are obtained from the gradient of the Hamiltonian [53]. Importantly, these calculations of gauge invariant quantities do not require a smooth Bloch gauge, and instead use the numerically determined eigenvectors. Figure 1 shows the gauge invariant structural quantities relevant for simulating the GI-SBEs. The minimum band gap is at $\Gamma (k = 0)$, where only the $c_1 - v$ TDM magnitude is nonvanishing. Figure 1(c) shows that all shift vectors exhibit zero crossings throughout the BZ, while in Fig. 1(d) we see that the triple product has a discontinuous phase jump at $\Gamma$ arising from a zero crossing of the complex TDMs.

The system is excited by a linearly polarized, 100 fs pulse with 2.07 $\mu$m wavelength and peak field strength 0.0042 a.u. using a dephasing time of 0.5 optical cycles. The simulated interband HHG spectra for the three-band model are shown in Fig. 2, with harmonic efficiency calculated as $S(\omega) = | \int dt e^{-i\omega t} j_{ct}(t) |^2$. Clearly, the GC-SBEs and GI-SBEs spectra in Fig. 2 are practically identical. Further, numerical integration of the GI-SBEs is unaffected by the nonsmooth behavior of the triple phase product.

In the following we demonstrate that the GI-SBEs are a useful tool to investigate the influence of the various band structure components. Figure 2(b) shows HHG in the two-band ($v$, $c_1$) limit, comparing $R_{c_1,v} = 0$ and $R_{c_1,v} \neq 0$. The shift vector is solely responsible for even-order HHG, in contrast to the GC-SBEs where the TDM phase and Berry connection account for symmetry breaking. Moreover, the shift vector barely influences odd harmonics. As a result, odd and even harmonics are driven by different band...
structural elements, absolute value of TDM, and shift vector, respectively, thus facilitating interpretation of HH spectra. We can also observe in Fig. 2(b) that the ratios between even and odd harmonics vary throughout the spectrum. For example, harmonic 14 is comparable in strength to harmonic 15, whereas harmonic 16 is much weaker than harmonic 17. This variation can be attributed to the \(k\)-dependent behavior of the shift vector; suppression of even harmonics happens when HHG takes place around \(k\) points in the BZ, where the shift vector vanishes; see Fig. 1(c).

We can gain further insight by comparing HH spectra calculated within different approximations. Figure 3(a) reveals the contributions from the \(c_2\) band by comparing two-band to three-band simulations. The region of largest discrepancy occurs near harmonic 19, but away from this region the two-band approximation works well. For more quantitative conclusions, we define a relative discrepancy of the harmonic spectrum for different simulation cases,

\[
\Delta S(\omega) = |S(\omega) - S^N(\omega)| / [S(\omega) + S^N(\omega)],
\]

where \(S^N\) is the spectrum from an approximate simulation labeled by \(N\). The HH discrepancies for three different approximations are shown in Fig. 3(b). The discrepancy for the two-band approximation is largest for harmonics 19–25; in agreement with Fig. 3(a). To uncover the influence of \(c_2\), starting from the two-band limit we first include the TDM magnitude \(|d_{c_2,v}|\) in case II, which significantly reduces the discrepancy of the odd harmonic orders. The discrepancy of the even harmonic orders is reduced by including the shift vector \(R_{c_2,v}\) in case III. From this we can draw two conclusions: (i) \(|d_{c_2,v}|\) and \(R_{c_2,v}\) are needed to model odd and even harmonics 19–25, respectively; and (ii) neither the \(c_1\)-\(c_2\) transition nor the triple product contribute significantly for these simulation conditions.

Next we use the SBEs to simulate HHG in the Haldane model for a Chern insulator on the 2D hexagonal lattice [59]. We simulate the Haldane model in its topological phase, using model parameters from Ref. [60] to obtain a minimum band gap of \(\sim 3\) eV. We use a linearly polarized 3.25 \(\mu\)m pulse with 50 fs duration and peak field strength 0.0045 a.u. Expressions for the band structure are provided in the SM [53]. Figures 4(a) and 4(b) show the shift vector corresponding to \(\Gamma K (\Gamma M)\) orientation of the linearly polarized driving laser. Simulated HHG spectra for the two orientations are shown in Figs. 4(c) and 4(d). Although the shift vectors exhibit rapidly varying behavior near \(\Gamma\), HH spectra calculated using the GI-SBEs are found to converge using a Monkhorst-Pack mesh of \(300 \times 300\) points. In contrast, the GC-SBEs involve Berry connections which are singular at the \(K\) points of the BZ, and they failed to converge using a denser grid of \(800 \times 800\) points. As such, the GI-SBEs can be integrated directly, while more elaborate techniques have to be used to improve convergence of the GC-SBEs [43,60].

Finally, we remark that the topology of a Chern insulator can be revealed directly from the shift vector. Referring to
Eq. (7), it is clear that the curl of the shift vector is simply the difference of Berry curvatures between two bands. As a result, \( \frac{1}{2\pi} \int_{BZ} d\mathbf{k} \nabla_{\mathbf{k}} \times \mathbf{R}_{\mathbf{k}}(\mathbf{k}) \) yields a topological invariant, namely, the difference of Chern numbers between bands, which can be either 0 or \( \pm 2 \), in the trivial and topological phase, respectively.

In conclusion, we have introduced the GI-SBEs which are formulated in terms of gauge invariant, physically measurable quantities. Both the GI-SBEs and the GC-SBEs are exact and yield the same results for physical observables. We have numerically validated the GI-SBEs and demonstrated their usefulness. The GI-SBEs were shown to provide a distinct advantage for a Chern insulator, in which the Berry connections have a nonremovable singularity due to topological obstruction of the Bloch phase. The GI-SBEs replace the Berry connection with the shift vector, which is independent of Bloch phase. Compared to the GC-SBEs, this leads to superior convergence and stability in numerical modeling, as demonstrated in our calculations.

Since the GI-SBEs involve only gauge invariant quantities, they provide a novel approach for reconstruction of band structure from HH spectra. Reconstruction has already been demonstrated for the gauge invariant Berry band structure from HH spectra. Reconstruction has quantities, they provide a novel approach for reconstruction of

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