Imperfect Recollisions in High-Harmonic Generation in Solids

Lun Yue and Mette B. Gaarde

1Department of Physics and Astronomy, Louisiana State University, Baton Rouge, Louisiana 70803-4001, USA

(Dated: February 5, 2020)

We theoretically investigate high-harmonic generation in hexagonal boron nitride with linearly polarized laser pulses. We show that imperfect recollisions between electron-hole pairs in the crystal give rise to an electron-hole-pair polarization energy that leads to a double-peak structure in the subcycle emission profiles. An extended recollision model (ERM) is developed that allows for such imperfect recollisions, as well as effects related to Berry connections, Berry curvatures, and transition-dipole phases. The ERM illuminates the distinct spectrototemporal characteristics of harmonics emitted parallel and perpendicularly to the laser polarization direction. Imperfect recollisions are a general phenomenon and a manifestation of the spatially delocalized nature of the real-space wave packet, they arise naturally in systems with large Berry curvatures, or in any system driven by elliptically polarized light.

The last decade has seen the emergence of high-harmonic generation (HHG) in solids \cite{1} as a promising and compact ultrafast light source, as well as a potential tool to reconstruct crystal band structures \cite{7}, measure Berry curvatures \cite{8,9}, and probe topological phase transitions \cite{10,12}. Complementing experimental progress, a number of theoretical studies have explored HHG in solids either in terms of reciprocal-space dynamics within the band structure \cite{1,5,6,13,23}, which contains both intra- and interband contributions, or in terms of real-space particle-like dynamics in the crystal \cite{2,3,14,24-29}.

The semi-classical recollision model, extended to solids \cite{14,24} from the gas-phase three-step model of strong-field interactions \cite{30,31}, has in particular provided an intuitive understanding of the interband contribution to HHG in solids: in each laser half-cycle, an electron tunnels from the valence band to the conduction band and leaves behind a hole; the laser field spatially drives the electron and hole according to their respective band-structure dispersions; when the electron and hole recollide, a high-energy photon is emitted with energy corresponding to the instantaneous band gap. At the recollision step, the assumption has been that the electron and hole reencounter each other exactly at the same spatial position. However, since Bloch waves are spatially delocalized, it is possible for the electron and hole wavepackets to spatially overlap even when their centers do not [Fig. 1(a)], with the implication that imperfect recollisions manifest themselves in the HHG process in solids, and provide an extended recollision model (ERM) that naturally includes this effect, as well as other properties such as the Berry connections and the TDPs. As a concrete example, we study HHG in monolayer hexagonal boron nitride (h-BN) driven by infrared pulses linearly polarized in the crystal plane, and we compare the ERM results to solutions of the semiconductor Bloch equations (SBEs) \cite{33,34}. In addition to the recent intense interest in HHG from two-dimensional materials \cite{8,28,35-39}, h-BN is interesting due to its lack of an inversion center which leads to non-zero Berry connections and TDPs. Also, pulse propagation effects \cite{10,11} can be neglected in monolayer materials. We find that when the driving laser is polarized along the $\Gamma - K$ direction, the eh-PER manifests itself in the time-frequency spectrograms of the parallel-emitted odd-order harmonics as a double-peak structure. Within the ERM this is explained as two sets of trajectories launched each half-cycle, from different $k$-points in the Brillouin zone (BZ). We show that the quantum interference between multiple $k$-point

![FIG. 1. (a) An imperfect recollision in which the electron and hole centers do not exactly overlap, leading to an electron-hole-pair polarization energy upon recollision. In the ERM, we also keep track of the phase accumulated along the trajectories. (b) Band gap energy (in units of $\omega_0 = 0.0285$) of monolayer h-BN with annotated points of interest. The gray discs around $M_1$ and $M_2$ each have radius 0.1.](image-url)
the band gap, $F$, inclusion of one valence and one conduction band, using solids and could potentially stimulate new experiments. We start by solving the SBEs for monolayer h-BN with inclusion of one valence and one conduction band, using a dephasing time $T_2 = 5$ fs. The band structure of monolayer h-BN [Fig. 1(b)] is obtained by the pseudopotential method of Ref. [42] and is given in the Supplemental Material (SM) [43]. The Berry connections and TDPs are calculated numerically in the twisted parallel transport gauge [44], ensuring continuity and BZ-periodicity. For the laser parameters in this paper, the HHG spectrum is converged with respect to increasing the number of bands. The filled curves in Figs. 2(a) and 2(b) show the high-harmonic spectrum of monolayer h-BN driven by a 1600 nm, 35 fs laser pulse with a peak intensity of 3.5 TW/cm². Harmonics up to 25th order are generated, with purely odd (even) orders generated parallel (perpendicular) to the LPD. The dotted lines in Figs. 2(a) and 2(b) show that inclusion of only the TDPs without the Berry connections, as e.g. was done in [32] to show the emergence of even harmonics in ZnO, would produce erroneous spectra.

Figures 2(c) and 2(d) show the sub-cycle time-frequency profiles for the emission with parallel and perpendicular ($\parallel$, $\perp$) polarization, respectively, and illustrate a key result of this paper: The parallel emission profile clearly exhibits a broad, double-peaked structure, whereas the perpendicular profile is single-peaked and much narrower, and in fact exhibits a pronounced minimum at the position corresponding to the second peak in the parallel emission.

To get a clear physical understanding of the emission dynamics observed in Fig. 2 we develop an extended version of the recollision model for HHG in solids [14, 24]. We focus on the interband dynamics which strongly dominates the emission above the band gap. Atomic units are used throughout this work unless indicated otherwise. In the two-band approximation and assuming a small population in the conduction band, the interband spectrum parallel and perpendicular ($\mu = \parallel, \perp$) to the LPD are

$$j_\mu(t) = \sum_k R_k^\mu \int_t^\infty T^\mu(s) e^{-i S_\mu(k,t,s)} ds + c.c.$$  \hspace{1cm} (1)

with $T^\mu(s) = F(s)\omega_\mu^\mu(s)$ the transition matrix element, $R_k^\mu = \omega_\mu^\mu |\alpha_k^\mu|$ the recombination dipole, $\omega_\mu^\mu = E_c^\mu - E_v^\mu$ the band gap, $F(t) = F(t)\hat{e}_\parallel$ the electric field with $\hat{e}_\parallel$ the laser polarization direction (LPD), when the Berry connections and TDPs are included. We find that the imperfect recollisions involve tens of a.u. separations at the time of recollision, and that this is consistent with the size of the delocalized quantum wave packet. The formulation of the ERM and in particular the inclusion of the eh-PER provides new insights into the HHG process in solids and could potentially stimulate new experiments.

We solve the SBEs for the high-harmonic generation (HHG) spectrum, the recombination dipole, and the Berry connections and TDPs are included. We find that the imperfect recollisions involve tens of a.u. separations at the time of recollision, and that this is consistent with the size of the delocalized quantum wave packet. The formulation of the ERM and in particular the inclusion of the eh-PER provides new insights into the HHG process in solids and could potentially stimulate new experiments.

Fig. 2. HHG spectra (a,b) and time-frequency profiles (c,d) for LPD along the $\Gamma - K$ direction, and high harmonics polarized parallel $(a,c)$ and perpendicular $(b,d)$ to the LPD. In (a) and (b), the filled curve results from the full SBE calculation, the dotted line neglects the Berry connections, and the dashed horizontal line outlines the minimal band gap. In (c) and (d), the color map is the quantum result; the lines and dots are semiclassical results, with tunneling from different symmetry points in the BZ [see Fig. 1(b)]. The red cross in (d) marks a minimum in the color map.

The Berry connections and TDPs are calculated numerically in the twisted parallel transport gauge [44], ensuring continuity and BZ-periodicity. For the laser parameters in this paper, the HHG spectrum is converged with respect to increasing the number of bands. The filled curves in Figs. 2(a) and 2(b) show the high-harmonic spectrum of monolayer h-BN driven by a 1600 nm, 35 fs laser pulse with a peak intensity of 3.5 TW/cm². Harmonics up to 25th order are generated, with purely odd (even) orders generated parallel (perpendicular) to the LPD. The dotted lines in Figs. 2(a) and 2(b) show that inclusion of only the TDPs without the Berry connections, as e.g. was done in [32] to show the emergence of even harmonics in ZnO, would produce erroneous spectra.

Figures 2(c) and 2(d) show the sub-cycle time-frequency profiles for the emission with parallel and perpendicular polarization, respectively, and illustrate a key result of this paper: The parallel emission profile clearly exhibits a broad, double-peaked structure, whereas the perpendicular profile is single-peaked and much narrower, and in fact exhibits a pronounced minimum at the position corresponding to the second peak in the parallel emission.

To get a clear physical understanding of the emission dynamics observed in Fig. 2 we develop an extended version of the recollision model for HHG in solids [14, 24]. We focus on the interband dynamics which strongly dominates the emission above the band gap. Atomic units are used throughout this work unless indicated otherwise. In the two-band approximation and assuming a small population in the conduction band, the interband spectrum parallel and perpendicular ($\mu = \parallel, \perp$) to the LPD are

$$j_\mu(t) = \sum_k R_k^\mu \int_t^\infty T^\mu(s) e^{-i S_\mu(k,t,s)} ds + c.c.$$  \hspace{1cm} (1)

The LPD, $d_k^\mu = i\langle u^n_k | \nabla_k | u^k_v \rangle$ the coupling matrix elements with $u^n_k$ the periodic part of the Bloch function, and $\kappa(\tau) = \mathbf{k} - \mathbf{A}(\tau) + \mathbf{A}(\tau)$ with $\mathbf{A}$ the vector potential satisfying $-dA/dt = \mathbf{F}$. The accumulated phase in Eq. (1) is (dephasing ignored)

$$S_\mu(k,t,s) = \int_s^\infty \left[ \omega_\mu^\mu(\tau) + \mathbf{F}(\tau) \cdot \nabla\kappa(\tau) \right] d\tau$$

$$+ \alpha_k^\mu - \alpha_\mu^\mu(s)$$  \hspace{1cm} (2)

with $\Delta\mathbf{A}^\mu = \mathbf{A}^\mu - \mathbf{A}^\mu_0$ where $\mathbf{A}^\mu_0 = i\langle u^n_k | \nabla_k | u^k_v \rangle$ are the Berry connections, and $\alpha_k^\mu = -\arg(d_k^\mu)$ the transition dipole phases (TDPs). The saddle point conditions for $S_\mu(k,t,s) - \omega t$ are

$$\omega_\mu^\mu(\tau) + \mathbf{F}(\tau) \cdot \nabla\kappa(\tau) = 0,$$  \hspace{1cm} (3a)

$$\Delta R_\mu \equiv \Delta r - d_\mu^\mu + D_\mu^\mu = 0,$$  \hspace{1cm} (3b)

$$\omega_\mu^\mu + \mathbf{F}(\tau) \cdot \left[ D^{\mu(s)} + \Delta r \right] = \omega,$$  \hspace{1cm} (3c)

with $D_\mu^\mu = \Delta\mathbf{A}^\mu - \nabla_k \alpha_k^\mu$ and $\Delta r \equiv \int_s^\tau \left[ v^n_c(\tau) - v^n_v(\tau) \right] d\tau$ where $v^n_c(\tau) \equiv \nabla_k E^n_c(\tau) + \mathbf{F}(\tau) \times \Omega^n_c(\tau)$ is the velocity including the Berry curvature $\Omega^n_k = \nabla_k \times \mathbf{A}^\mu_0$. Semiclassically, Eqs. (3a), (3c) are interpreted in terms of the three steps in the recollision model: at time $s$, an electron tunnels from the valence to the conduction band with crystal
harmonics with orders \(\gtrsim\) seen here to be responsible for the emitted high-order collisions distance \(\Delta r\). The M\(_1\) energies are plotted for different initial crystal momenta and energies only peaks once during each half-cycle (with each density of states diverging at the M points in h-BN \([47]\). Our ERM \([45]\) extends previous works by including (i) \(\omega\) crystal momentum \(k\) accelerated by the laser and recollides at time \(t\) \(\equiv 0\). In Figs. 2(c) and 2(d), the semiclassical recollision energies versus recollision times (a) without and (b) with inclusion of the eh-PER \(F \cdot \Delta r\) in Eq. (3c), for LPD along \(\Gamma - K\). The different curves correspond to different tunneling sites in the BZ [Fig. 1(b)]. The vertical dotted lines mark the peaks of M\(_1\) and M\(_2\) recollision energies. The inset in (b) shows the spatial spread corresponding to the trajectory that tunnels at M\(_1\) with the highest recollision energy. The shaded curves in (c,d) show the quantum interferences between the M\(_1\) and M\(_2\) recollision energies. The inset in (b) shows the spatial spread corresponding to different tunneling sites in the BZ [Fig. 1(b)]. The different curves correspond to different tunneling sites in the BZ [Fig. 1(b)]. The different curves correspond to different tunneling sites in the BZ [Fig. 1(b)].

As seen in Figs. 3(a) and 3(b), both the recollision times and the recollision energies are modified when taking into account the eh-PERs: M\(_1\) and M\(_2\) (M\(_1\) and M\(_2\)) recollide later (earlier with higher energy) during the first half-cycle, and earlier with higher energy (later) during the second half-cycle. Henceforth, we will refer to the two peaks during each half-cycle as early and late emissions. In Figs. 3(a) and 3(b), we have only considered the \(\mu = \parallel\) case; for \(\mu = \perp\), the result is similar, i.e. with a double-peak structure in the semiclassical recollision energies when including the eh-PER.

Note that our choice \(R_0 = 30\) is several times greater than the h-BN lattice constant of 4.7, stressing the delocalized character of the spatial recollision process. When decreasing \(R_0\), the maximum recollision times of the M\(_1\) and M\(_1\) curves in Fig. 2(b) become shorter, with the peak structures, and thereby all “long” trajectories, disappearing around \(R_0 \sim 20\) (see SM). For the electron and hole wave packets to spatially overlap at recollision time, the good agreement between our semiclassical and quantum results [Fig. 3(b) and Fig. 2(c)] thus suggests that the quantum wave packet has a minimum spread of \(\sim 30\). We have further estimated the quantum spatial spread by explicitly constructing a real-space wave packet during time propagation, after placing a \(k\)-space wave packet on the conduction band (using the Houston-state basis \([50, 51]\) with a \(k\)-width estimated by tunneling (see SM for more details). For the trajectory that tunnels at the M\(_1\) point and has the highest recollision energy, the spatial spread \(\sigma\) of the corresponding wave packet is shown in the inset of Fig. 3(b), where \(\sigma\) is seen to increase from 39 to 48 between tunnel and recollision. These \(\sigma\) values are fully
consistent with our choice of $R_0$ and the previous discussion. The double-peak structure is also robust with respect to the choice of $T_2$, even up to 20 fs (see SM).

If the recollision model predicts a double-peak structure for both the parallel and perpendicular time-profiles, why do we not see this in the quantum result of Fig. 2(d)? The answer lies in the quantum phase information of the trajectories. If two trajectories tunnel from two different $k$-points in the BZ, $P_1$ and $P_2$, and have the same recollision energy at the same recollision time $t$, we can coherently add them as

$$P_{P_1P_2}(t) = \frac{1}{2} \left[ e^{iS_{P_1}(t)} + e^{iS_{P_2}(t)} \right],$$

with $S_{P_1}(t)$ and $S_{P_2}(t)$ the accumulated phases [Eq. (2)].

For the parallel harmonics in Fig. 3(c), $|M_1M_2(t)| = 1$ for all $t$, indicating that the emission from $M_1$ and $M_2$ is completely in-phase. In contrast, for the perpendicular case in Fig. 3(d), the late (early) emissions exhibit clear destructive (constructive) interferences. In addition, the pronounced minimum in the perpendicular time-frequency profile in Fig. 2(d) is exactly reproduced by the interference minimum in Fig. 3(d) shown by the red cross. Thus with our ERM, we can even explain quantitative details in the emission profiles. Note that the difference in the accumulated phases between the parallel and perpendicular harmonics can be traced back to the TDPs $\alpha_{P_1}$ in Eq. (2), stressing their importance for HHG in solids. Similarly, within our framework, the generation of purely odd (even) harmonics in the parallel (perpendicular) directions shown in Fig. 2(a) [Fig. 2(b)] is easily explained by considering interference between emissions originating in tunneling from the $M_1$ and $M_2$ sites, as illustrated in Fig. 4. For the parallel polarization, the condition $I_{M_1M_2}(t) = -I_{M_1M_2}(t + \frac{T}{2})$ leads to purely odd harmonics, whereas for the perpendicular polarization, only even harmonics are generated due to $I_{M_1M_2}(t) = I_{M_1M_2}(t + \frac{T}{2})$.

Finally, we discuss the general situations in which imperfect recollisions should be important for HHG in solids. Clearly, the eh-PER contribution to the harmonic energy is ubiquitous in all solids where the electron and hole do not exactly spatially recollide. Obvious examples include HHG in solids with elliptical drivers, and systems with large Berry curvatures. For linear polarization, the general rule is the following: suppose an electron-hole-pair is created at a symmetry point $S_0$ and subsequently driven by a LD along $\Gamma - K$, then the eh-PER will be important when $S \neq S_0$. This is illustrated in Fig. 5 where for the $\Gamma - K$ polarization, the excursions of the electron-hole-pairs created at $M_1$ and $M_2$ in $k$-space, $k(t)$ (teal arrows), are not along the group velocity vector fields $\nabla_k E_n$, yielding a nonzero eh-PER. In contrast, when the LD is $\Gamma - M_1$, the $k$-space excursion (purple arrows) are along the group velocities, such that the electron and hole travel along a straight line in real space, leading to perfect recollisions with $\Delta r = 0$. Indeed, this is confirmed by both our quantum and semiclassical calculations in the inset of Fig. 5 where no double peak is observed and all trajectories starting from a $M_1$ disc in the BZ have approximately the same emission times. We emphasize that the double-peak time profiles originate in emission from different $k$-points in the BZ with similar emission times and we therefore do not expect them to be significantly altered by macroscopic effects such as phase matching or intensity averaging [40]. Indeed, we checked that the emission structures remain even when using $T_2 = 2$ fs in the SBEs, and are robust with respect to focal-volume averaging and spatial filtering in the far field [52].

In conclusion, we have uncovered and characterized the
effects of the imperfect recollisions for HHG in solids. In the example system of h-BN, they manifest in the time-frequency profiles as a double peak structure in the sub-cycle emission profile of the parallel-polarized harmonics, which is absent in the perpendicular-polarized emission. In the process, we formulated an ERM for HHG in solids that captures the effects of the eh-PER, as well as the temporal dephasing introduced by e.g. electron correlation. We predict that eh-PER should be ubiquitous in a large range of extreme nonlinear phenomena of current interest, such as HHG with elliptical drivers and systems with large Berry curvatures, as well as high-order sideband generation. The identification of eh-PER and its effect on the harmonic emission profiles, as well as the formulation of the ERM, provides new insights into the HHG process in solids and could potentially stimulate new experiments. The potential experimental measurement of such subcycle emission dynamics could also give us information on where in the BZ the trajectories emanate, thus probing the dynamical band structures. More generally, the characterization and understanding of harmonic emission times and spectral phases are very important for attosecond metrology in solids.

The authors acknowledge support from the National Science Foundation, under Grant No. PHY1713671. L. Y. gratefully acknowledges Francois Mauger and Kenneth J. Schafer for useful discussions.

[1] S. Ghimire, A. D. DiChiara, E. Sistrunk, P. Agostini, L. F. DiMauro, and D. A. Reis, “Observation of high-order harmonic generation in a bulk crystal,” Nat. Phys. 7, 138–141 (2011).

[2] G. Vampa, T. J. Hammond, N. Thiré, B. E. Schmidt, F. Légaré, C. R. McDonald, T. Brabec, and P. B. Corkum, “Linking high harmonics from gases and solids,” Nature 522, 462 EP – (2015).

[3] Y. S. You, D. A. Reis, and S. Ghimire, “Anisotropic high-harmonic generation in bulk crystals,” Nat. Phys. 13, 345 EP – (2016).

[4] G. Ndagbashimiye, S. Ghimire, M. Wu, D. A. Browne, K. J. Schafer, M. B. Gaarde, and D. A. Reis, “Solid-state harmonics beyond the atomic limit,” Nature 534, 520 EP – (2016).

[5] M. Garg, M. Zhan, T. T. Luu, H. Lakhota, T. Klostermann, A. Guggenmos, and E. Goulielmakis, “Multi-petahertz electronic metrology,” Nature 538, 359 EP – (2016).

[6] Z. Wang, H. Park, Y. H. Lai, J. Xu, C. I. Blaga, F. Yang, P. Agostini, and L. F. DiMauro, “The roles of photocarrier doping and driving wavelength in high harmonic generation from a semiconductor,” Nat. Commun. 8, 1686 (2017).

[7] G. Vampa, T. J. Hammond, N. Thiré, B. E. Schmidt, F. Légaré, C. R. McDonald, T. Brabec, D. D. Klug, and P. B. Corkum, “All-optical reconstruction of crystal band structure,” Phys. Rev. Lett. 115, 193603 (2015).

[8] H. Liu, Y. Li, Y. S. You, S. Ghimire, T. F. Heinz, and D. A. Reis, “High-harmonic generation from an atomically thin semiconductor,” Nat. Phys. 13, 262 EP – (2017).

[9] T. T. Luu and H. J. Wörner, “Measurement of the berry curvature of solids using high-harmonic spectroscopy,” Nat. Commun. 9, 916 (2018).

[10] D. Bauer and K. K. Hansen, “High-harmonic generation in solids with and without topological edge states,” Phys. Rev. Lett. 120, 177401 (2018).

[11] R. E. F. Silva, A. Jiménez-Galán, B. Amorim, O. Smirnova, and M. Ivanov, “Topological strong-field physics on sub-laser-cycle timescale,” Nat. Photonics 13, 849 (2019).

[12] A. Chacón, W. Zhu, S. P. Kelly, A. Dauphin, E. Pisanty, A. Picón, C. Ticknor, M. F. Ciappina, A. Saxena, and M. Lewenstein, “Observing topological phase transitions with high harmonic generation,” arXiv:1807.01616v2 (2019).

[13] A. F. Kemper, B. Moritz, J. K. Freericks, and T. P. Devereaux, “Theoretical description of high-order harmonic generation in solids,” New Journal of Physics 15, 023003 (2013).

[14] G. Vampa, C. R. McDonald, G. Orlando, D. D. Klug, P. B. Corkum, and T. Brabec, “Theoretical analysis of high-harmonic generation in solids,” Phys. Rev. Lett. 113, 073901 (2014).

[15] O. Schubert, M. Hohenleutner, F. Langer, B. Urbanek, C. Lange, U. Huttner, D. Golde, T. Meier, M. Kira, S. W. Koch, and R. Huber, “Sub-cycle control of terahertz high-harmonic generation by dynamical bloch oscillations,” Nat. Photonics 8, 119 (2014).

[16] T. Higuchi, M. I. Stockman, and P. Hommelhoff, “Strong-field perspective on high-harmonic radiation from bulk solids,” Phys. Rev. Lett. 113, 213901 (2014).

[17] T. T. Luu, M. Garg, S. Y. Kruchinin, A. Moulet, M. T. Hassan, and E. Goulielmakis, “Extreme ultraviolet high-harmonic spectroscopy of solids,” Nature 521, 498 EP – (2015).

[18] M. Wu, S. Ghimire, D. A. Reis, K. J. Schafer, and M. B. Gaarde, “High-harmonic generation from bloch electrons in solids,” Phys. Rev. A 91, 043839 (2015).

[19] M. Hohenleutner, F. Langer, O. Schubert, M. Knorr, U. Huttner, S. W. Koch, M. Kira, and R. Huber, “Real-time observation of interfering crystal electrons in high-harmonic generation,” Nature 523, 572 EP – (2015).

[20] T. Tamaya, A. Ishikawa, T. Ogawa, and K. Tanaka, “Diabatic mechanisms of higher-order harmonic generation in solid-state materials under high-intensity electric fields,” Phys. Rev. Lett. 116, 016601 (2016).

[21] M. Garg, H. Y. Kim, and E. Goulielmakis, “Ultimate waveform reproducibility of extreme-ultraviolet pulses by
high-harmonic generation in quartz,” Nat. Photonics 12, 291–296 (2018).

[22] L. Li, P. Lan, X. Zhu, T. Huang, Q. Zhang, M. Lein, and P. Lu, “Reciprocal-space-trajectory perspective on high-harmonic generation in solids,” Phys. Rev. Lett. 122, 193901 (2019).

[23] P. Navarrete, M. F. Ciappina, and U. Thumm, “Crystal-momentum-resolved contributions to high-order harmonic generation in solids,” Phys. Rev. A 100, 033405 (2019).

[24] G. Vampa, C. R. McDonald, G. Orlando, P. B. Corkum, and T. Brabec, “Semiclassical analysis of high harmonic generation in bulk crystals,” Phys. Rev. B 91, 064302 (2015).

[25] C. Yu, K. K. Hansen, and L. B. Madsen, “Enhanced high-order harmonic generation in donor-doped band-gap materials,” Phys. Rev. A 99, 013435 (2019).

[26] T. Ikemachi, Y. Shinohara, T. Sato, J. Yumoto, W. V. Houston, “Acceleration of electrons in a crystal lattice,” Phys. Rev. 57, 184–186 (1940).

[27] M. A. Denecke, “Nonperturbative harmonic generation in solids,” Phys. Rev. A 95, 033411 (2017).

[28] N. Tancogne-Dejean and O. D. Mücke, “Ellipticity dependence of high-harmonic generation in solids originating from coupled intraband and interband dynamics,” Nat. Commun. 8, 745 (2017).

[29] N. Tancogne-Dejean and A. Rubio, “Atomic-like high-harmonic generation from two-dimensional materials,” Sci. Adv. 4 (2018), 10.1126/sciadv.aao5207.

[30] N. Yoshikawa, K. Nagai, K. Uchida, Y. Takaguchi, S. Sasaki, Y. Miyata, and K. Tanaka, “Interband resonant high-harmonic generation by valley polarized electron-hole pairs,” Nat. Commun. 10, 3709 (2019).

[31] P. B. Corkum, “Plasma perspective on strong field multiphoton ionization,” Phys. Rev. Lett. 71, 1994–1997 (1993).

[32] M. Lewenstein, P. Balcou, M. Y. Ivanov, A. L’Huillier, and P. B. Corkum, “Theory of high-harmonic generation by low-frequency laser fields,” Phys. Rev. A 49, 2117 (1994).

[33] S. Jiang, J. Chen, H. Wei, C. Yu, R. Lu, and C. D. Lin, “Role of the transition dipole amplitude and phase on the generation of odd and even high-order harmonics in crystals,” Phys. Rev. Lett. 120, 253201 (2018).

[34] D. Golde, T. Meier, and S. W. Koch, “High harmonics generated in semiconductor nanostructures by the coupled dynamics of optical inter- and intraband excitations,” Phys. Rev. B 77, 075330 (2008).

[35] M. Kira and S. W. Koch, Semiconductor Quantum Optics (Cambridge University Press, 2012).

[36] N. Yoshikawa, T. Tamaya, and K. Tanaka, “High-harmonic generation in graphene enhanced by elliptically polarized light excitation,” Science 356, 736–738 (2017).

[37] M. Taucer, T. J. Hammond, P. B. Corkum, G. Vampa, C. Couture, N. Thiré, B. E. Schmidt, F. Légaré, H. Selvi, N. Unseree, B. Hamilton, T. J. Echtmaner, and M. A. Denecke, “Nonperturbative harmonic generation in graphene from intense midinfrared pulsed light,” Phys. Rev. B 96, 195420 (2017).

[38] H. A. Hláøz, S. Kovalev, J.-C. Deinert, Z. Mics, B. Green, N. Avari, M. Chen, S. Germanuskii, U. Lehner, J. Teichert, Z. Wang, K.-J. Tielrooij, Z. Liu, Z. Chen, A. Narita, K. Müllen, M. Bonn, M. Gesch, and D. Turchinovich, “Extremely efficient terahertz high-harmonic generation in graphene by hot dirac fermions,” Nature 561, 507–511 (2018).

[39] R. Silva, F. Martín, and M. Ivanov, “High harmonic generation in crystals using maximally localized wannier functions,” arXiv:1904.00283v2 (2019).

[40] M. S. Mrudul, N. Tancogne-Dejean, A. Rubio, and G. Dixit, “High-harmonic generation from spin-polarised defects in solids,” arXiv:1906.10224 (2019).

[41] I. Floss, C. Lemell, G. Wachter, V. Smekal, S. A. Sato, X.-M. Tong, K. Yabana, and J. Burgdörfer, “Ab initio multiscale simulation of high-order harmonic generation in solids,” Phys. Rev. A 97, 011401(R) (2018).

[42] J. Lu, E. F. Cunningham, Y. S. You, D. A. Reis, and S. Ghimire, “Interferometry of dipole phase in high harmonics from solids,” Nat. Photonics 13, 96 (2019).

[43] A. Taghizadeh, F. Hipolito, and T. G. Pedersen, “Linear and nonlinear optical response of crystals using length and velocity gauges: Effect of basis truncation,” Phys. Rev. B 96, 195413 (2017).

[44] See Supplemental Material at [URL] for additional details about the structure calculations, the SBEs, the choice of $R_0$, and the method used in the calculation of the quantum wave packet spreading.

[45] D. Vanderbilt, Berry phases in electronic structure theory: electric polarization, orbital magnetization and topological insulators (Cambridge University Press, 2018).

[46] Note that similar equations appear in [53], but without inclusion of the electron-hole polarization energy. They were applied to a model one-dimensional system where the Berry connections can naturally chosen to be zero. We stress that imperfect recollisions can only occur in systems with dimensions higher than one.

[47] This is consistent with the discussion of laser dressing in Ref. 20.

[48] F. Hipolito, T. G. Pedersen, and V. M. Pereira, “Nonlinear photocurrents in two-dimensional systems based on graphene and boron nitride,” Phys. Rev. B 94, 045434 (2016).

[49] L. Van Hove, “The occurrence of singularities in the elastic frequency distribution of a crystal,” Phys. Rev. 89, 1189 (1953).

[50] A. J. Uzan, G. Orenstein, A. Jiménez-Galán, C. McDonald, R. E. F. Silva, B. D. Bruner, N. D. Klimkin, V. Blanchet, T. Arusi-Parpar, M. Krüger, A. N. Rubtsov, O. Smirnova, M. Ivanov, B. Yan, T. Brabec, and N. Dudovich, “Multi-band petahertz currents resolved via high harmonic generation spectroscopy,” arXiv:1904.00283v2 (2019).

[51] W. V. Houston, “Acceleration of electrons in a crystal lattice,” Phys. Rev. 57, 184–186 (1940).

[52] J. B. Krieger and G. J. Iafrate, “Time evolution of bloch electrons in a homogeneous electric field,” Phys. Rev. B 33, 5494 (1986).

[53] C. Q. Abadie, M. Wu, and M. B. Gaarde, “Spatiotemporal filtering of high harmonics in solids,” Opt. Lett. 43, 5339 (2018).

[54] T. Huang, X. Zhu, L. Li, X. Liu, P. Lan, and P. Lu, “High-order-harmonic generation of a doped semiconductor,” Phys. Rev. A 96, 043425 (2017).

[55] S. Almalki, A. M. Parks, G. Bart, P. B. Corkum, T. Brabec, and C. R. McDonald, “High harmonic generation tomography of impurities in solids: Conceptual analysis,” Phys. Rev. B 98, 144307 (2018).

[56] K. Chinzei and T. N. Ikeda, “Disorder effects on the origin of high-order harmonic generation in solids,” Phys.
[56] Oscar Zurrón-Cifuentes, R. Boyero-García, C. Hernández-García, A. Picón, and L. Plaja, “Optical anisotropy of non-perturbative high-order harmonic generation in gapless graphene,” Opt. Express 27, 7776–7786 (2019).

[57] X. Zhang, J. Li, Z. Zhou, S. Yue, H. Du, L. Fu, and H.-G. Luo, “Ellipticity dependence transition induced by dynamical bloch oscillations,” Phys. Rev. B 99, 014304 (2019).

[58] D. Xiao, M.-C. Chang, and Q. Niu, “Berry phase effects on electronic properties,” Rev. Mod. Phys. 82, 1959 (2010).

[59] B. Zaks, R. B. Liu, and M. S. Sherwin, “Experimental observation of electron-hole recollisions,” Nature 483, 580 (2012).

[60] F. Langer, M. Hohenleutner, C. P. Schmid, C. Poellmann, P. Nagler, T. Korn, C. Schüller, M. S. Sherwin, U. Huttner, J. T. Steiner, S. W. Koch, M. Kira, and R. Huber, “Lightwave-driven quasiparticle collisions on a subcycle timescale,” Nature 533, 225 EP – (2016).

[61] H. B. Banks, Q. Wu, D. C. Valovcin, S. Mack, A. C. Gossard, L. Pfeiffer, R.-B. Liu, and M. S. Sherwin, “Dynamical birefringence: Electron-hole recollisions as probes of berry curvature,” Phys. Rev. X 7, 041042 (2017).

[62] F. Langer, C. P. Schmid, S. Schlauderer, M. Gmitra, J. Fabian, P. Nagler, C. Schüller, T. Korn, P. G. Hawkins, J. T. Steiner, U. Huttner, S. W. Koch, M. Kira, and R. Huber, “Lightwave valleytronics in a monolayer of tungsten diselenide,” Nature 557, 76–80 (2018).

[63] J. Li, X. Zhang, S. Fu, Y. Feng, B. Hu, and H. Du, “Phase invariance of the semiconductor bloch equations,” Phys. Rev. A 100, 043404 (2019).