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Imperfect Recollisions in High-Harmonic Generation in Solids

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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 spectrotemporal 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 [1–6] as a promising and compact ultrafast light source, as well as a potential tool to reconstruct crystal band structures [7], measure Berry curvatures [8, 9], and probe topological phase transitions [10–12]. Complementing experimental progress, theoretical studies have explored HHG in solids either in terms of reciprocal-space dynamics within the band structure [1, 5, 6, 13–23], which contains both intra- and interband contributions, or in terms of real-space particle-like dynamics in the crystal [2, 3, 14, 24–29].

The semi-classical recollision model, extended to solids [14, 24] from the gas-phase three-step model of strong-field interactions [30, 31], has provided an intuitive real-space understanding of the interband contribution to HHG in solids: in each laser half-cycle, an electron tunnels from the valence 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, the electron and hole wavepackets can spatially overlap even when their centers do not [Fig. 1(a)], with the implication that an electron-hole-pair polarization energy at recollision (eh-PER) will contribute to the emitted photon energy. Also, the original recollision model does not discern between the parallel and perpendicularly polarized currents, and the omission of ubiquitous solid-state properties such as the Berry connection, Berry curvatures, and transition dipole phases (TDPs) are generally not well-justified. For instance, a recent numerical work [32] highlighted the importance of the TDPs on the generation of even-order harmonics in ZnO. It is thus desirable to construct a framework that includes all these afore-mentioned concepts.

In this Letter, we consider how imperfect recollisions

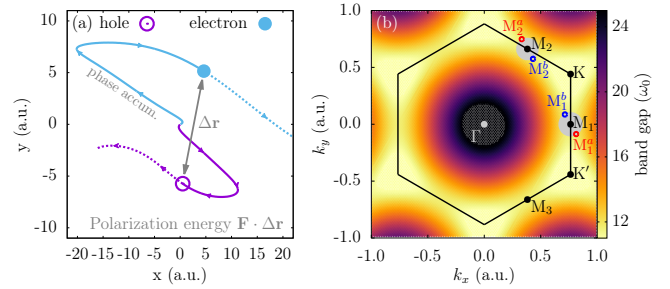


FIG. 1. (a) An imperfect recollision in which the electron and hole centers do not exactly overlap, leading to an electron-hole-pair polarization energy upon recollision. In the ERM, we also keep track of the phase accumulated along the trajectories. (b) Band gap energy (in units of $\omega_0 = 0.0285$) of monolayer h-BN with annotated points of interest. The gray discs around M_1 and M_2 each has radius 0.1.

manifest themselves in the HHG process in solids, and provide an extended recollision model (ERM) that naturally includes this effect and 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) [33, 34]. In addition to the recent intense interest in HHG from two-dimensional materials [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 [40, 41] can be neglected in monolayer materials. We find that when the driving laser is polarized along the Γ – 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 \mathbf{k} -points in the Brillouin zone (BZ). We show that the quantum interference between multiple \mathbf{k} -point contributions gives rise to different spectrotemporal characteristics of harmonic

emission parallel or perpendicular to the laser polarization direction (LPD), when the Berry connections and the 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 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 [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, 29.4 fs laser pulse with a peak intensity of 3.5 TW/cm^2 . 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 would produce erroneous spectra.

Figures 2(c) and 2(d) show the subcycle time-frequency emission profiles (EPs) with parallel and perpendicular polarization, respectively, and illustrate a key result of this paper: The parallel EP 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 is $\mathbf{j}(\omega) = \int_{-\infty}^{\infty} dt e^{i\omega t} \mathbf{j}(t)$, where the current components parallel and perpendicular ($\mu = \parallel, \perp$) to the LPD are

$$j_\mu(t) = \sum_{\mathbf{k}} R_\mu^{\mathbf{k}} \int_{-\infty}^t T^{\kappa(s)} e^{-iS_\mu(\mathbf{k}, t, s)} ds + \text{c.c.} \quad (1)$$

with $T^{\kappa(s)} = F(s)|d_\parallel^{\kappa(s)}|$ the transition matrix element, $R_\mu^{\mathbf{k}} = \omega_g^{\mathbf{k}}|d_\mu^{\mathbf{k}}|$ the recombination dipole, $\omega_g^{\mathbf{k}} = E_c^{\mathbf{k}} - E_v^{\mathbf{k}}$ the band gap, $\mathbf{F}(t) = F(t)\hat{\mathbf{e}}_\parallel$ the electric field with $\hat{\mathbf{e}}_\parallel$ the LPD, $\mathbf{d}^{\mathbf{k}} = i\langle u_c^{\mathbf{k}} | \nabla_{\mathbf{k}} | u_v^{\mathbf{k}} \rangle$ the coupling matrix elements with $u_m^{\mathbf{k}}$ the periodic part of the Bloch function, and $\kappa(\tau) = \mathbf{k} - \mathbf{A}(t) + \mathbf{A}(\tau)$ with \mathbf{A} the vector potential satisfying $-d\mathbf{A}/dt = \mathbf{F}$. The accumulated phase in

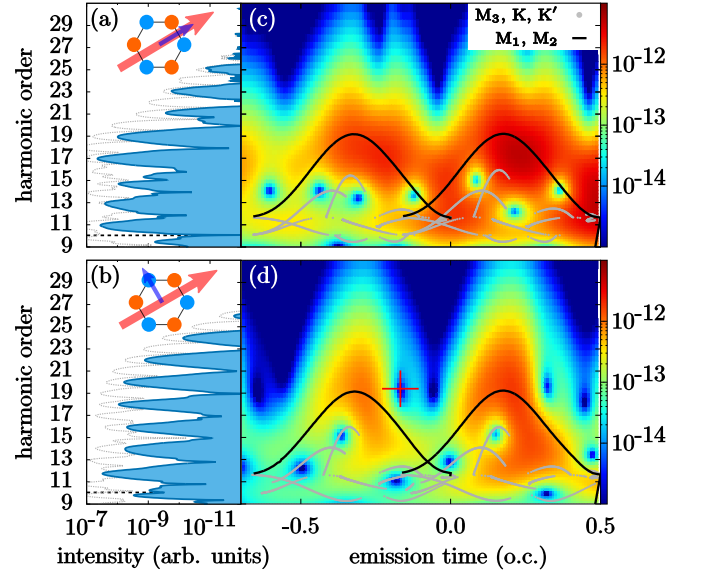


FIG. 2. HHG spectra (a,b) and EPs (c,d) for LPD along the $\Gamma - K$ direction, and high harmonics polarized parallel (a,c) and perpendicular (b,d) to the LPD. In (a) and (b), the filled curve results from the full SBE calculation, the dotted line neglects the Berry connections, and the dashed horizontal 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.

Eq. (1) is (dephasing ignored)

$$S_\mu(\mathbf{k}, t, s) = \int_s^t \left[\omega_g^{\kappa(\tau)} + \mathbf{F}(\tau) \cdot \Delta \mathcal{A}^{\kappa(\tau)} \right] d\tau + \alpha_\mu^{\mathbf{k}} - \alpha_\parallel^{\kappa(s)} \quad (2)$$

with $\Delta \mathcal{A}^{\mathbf{k}} = \mathcal{A}_c^{\mathbf{k}} - \mathcal{A}_v^{\mathbf{k}}$ where $\mathcal{A}_n^{\mathbf{k}} = i\langle u_n^{\mathbf{k}} | \nabla_{\mathbf{k}} | u_n^{\mathbf{k}} \rangle$ are the Berry connections, and $\alpha_\mu^{\mathbf{k}} = \arg(d_\mu^{\mathbf{k}})$ the transition dipole phases (TDPs). The saddle point conditions for $S_\mu(\mathbf{k}, t, s) - \omega t$ are

$$\omega_g^{\kappa(s)} + \mathbf{F}(s) \cdot \mathcal{D}_\parallel^{\kappa(s)} = 0, \quad (3a)$$

$$\Delta \mathbf{R}_\mu \equiv \Delta \mathbf{r} - \mathcal{D}_\mu^{\mathbf{k}} + \mathcal{D}_\parallel^{\kappa(s)} = 0, \quad (3b)$$

$$\omega_g^{\mathbf{k}} + \mathbf{F}(t) \cdot [\mathcal{D}_\parallel^{\kappa(s)} + \Delta \mathbf{r}] = \omega, \quad (3c)$$

with $\mathcal{D}_\mu^{\mathbf{k}} \equiv \Delta \mathcal{A}^{\mathbf{k}} - \nabla_{\mathbf{k}} \alpha_\mu^{\mathbf{k}}$ and $\Delta \mathbf{r} \equiv \int_s^t [\mathbf{v}_c^{\kappa(\tau)} - \mathbf{v}_v^{\kappa(\tau)}] d\tau$ where $\mathbf{v}_n^{\kappa(\tau)} \equiv \nabla_{\mathbf{k}} E_n^{\kappa(\tau)} + \mathbf{F}(\tau) \times \boldsymbol{\Omega}_n^{\kappa(\tau)}$ is the velocity including the Berry curvature $\boldsymbol{\Omega}_n^{\kappa(\tau)} \equiv \nabla_{\mathbf{k}} \times \mathcal{A}_n^{\kappa(\tau)}$. 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 momentum $\kappa(s)$; the newly created electron-hole pair is accelerated by the laser and recollides at time t with final crystal momentum \mathbf{k} and relative distance $\Delta \mathbf{r}$; at recollision, a high-energy photon with energy ω is released.

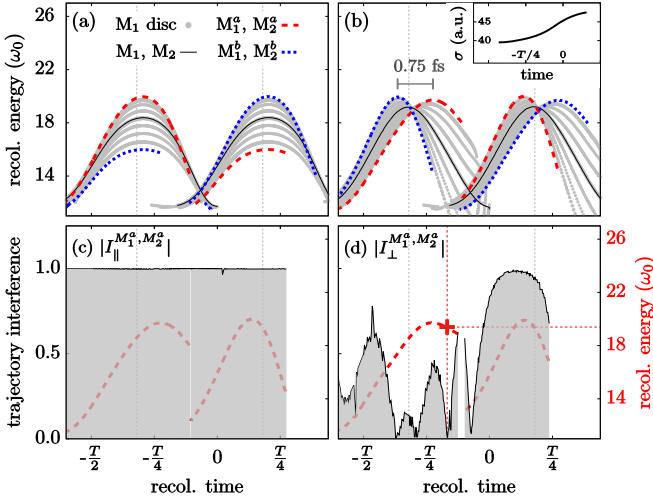


FIG. 3. Semiclassical recollision energies versus recollision times (a) without and (b) with inclusion of the eh-PER $\mathbf{F} \cdot \Delta \mathbf{r}$ in Eq. (3c), for LPD along $\Gamma - K$. The different curves correspond to different tunneling sites in the BZ [Fig. 1(b)]. The vertical dotted lines 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^a and M_2^a emissions, for the (c) parallel and (d) perpendicular directions (see text). The recollision energies (right y-axis) for M_1^a and M_2^a trajectories are redrawn in red dashed lines. In (d), the red cross indicates the same interference minimum as in Fig. 2(d).

Our ERM [45] extends previous works by including (i) laser-dressing of the bands with $\mathbf{F} \cdot \mathcal{D}_{\parallel}^{\kappa(s)}$; (ii) the Berry curvature contribution to the velocity of the trajectories; (iii) the eh-PER given by $\mathbf{F} \cdot \Delta \mathbf{r}$, due to the nonzero recollision distance $\Delta \mathbf{r} \neq \mathbf{0}$. In addition, we keep track of the accumulated phase of each trajectory. For h-BN and the field parameters used here, points (i) and (ii) are of minor importance [46]. In all our ERM calculations, a continuous-wave laser is employed, with tunneling times $s \in [-T, 0]$ where T is the period. Returning trajectories with $|\Delta \mathbf{R}_{\mu}| < R_0$ are assumed to have recollided, with $R_0 \equiv 30$ unless indicated otherwise.

Figures 2(c,d) show the semiclassical recollision energies for different initial crystal momenta $\kappa(s)$ corresponding to different symmetry points in the BZ [Fig. 1(b)]. The M_1 and M_2 points are seen here to be responsible for the emitted high-order harmonics with orders $\gtrsim 16$. This is consistent with the density of states diverging at the M points in h-BN [47] (van Hove singularities [48]), and strong HHG emissions are expected at such points [29, 49]. The recollision energy only peaks once during each half-cycle (with each energy below the maximum being emitted twice, resulting from a “short” and “long” trajectory, respectively, similar to gas-phase HHG), which differs from the double peak structure in the quantum result of Fig. 2(c). In addition, the recoli-

sion energies for the perpendicular harmonics are almost identical to those of the parallel case and do not reproduce the narrow SBE EP [Fig. 2(d)].

The imperfect recollisions, as we now demonstrate, are responsible for the double-peak structure in Fig. 2(c). Indeed, when we take into account eh-PER and all crystal momenta in a disc of radius 0.1 around the M_1 point in the BZ [henceforth referred to as the M_1 disc, see Fig. 1(b)], we recover the double peak structure in the semiclassical recollision model, shown in Fig. 3(b) by the gray dots. The time-delay between the two emissions is ~ 0.75 fs, with the first emission slightly higher in energy, in agreement with the quantum result of Fig. 2(c). When the eh-PERs are neglected in Fig. 3(a), the M_1 disc emits the highest-order harmonics at the same time, and no double-peak structure is observed. To better understand the role of the eh-PERs, we choose to consider two pairs of representative \mathbf{k} -points on the periphery of the M_1 and M_2 discs [red and blue circles in 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^a and M_2^a (M_1^b and M_2^b) 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^a and M_1^b curves in Fig. 3(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 ~ 30 . We have further estimated the quantum spatial spread by explicitly constructing a real-space wave packet during time propagation, after placing a \mathbf{k} -space wave packet on the conduction band (using the Houston-state basis [50, 51]) with a \mathbf{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 σ of the corresponding wave packet is shown in the inset of Fig. 3(b), where σ is seen to increase from 39 to 48 between tunnel and recollision. These σ 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). This is consistent with our previous comments: from the time of tunneling, the electron and hole wave packets are

driven apart spatially, allowing for recollision primarily during the first optical cycle of the pulse.

If the recollision model predicts a double-peak structure for both the parallel and perpendicular EP, 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 \mathbf{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

$$I_{\mu}^{P_1 P_2}(t) = \frac{1}{2} \left[e^{S_{\mu}^{P_1}(t)} + e^{S_{\mu}^{P_2}(t)} \right], \quad (4)$$

with $S_{\mu}^{P_1}(t)$ and $S_{\mu}^{P_2}(t)$ the accumulated phases [Eq. (2)]. For the parallel harmonics in Fig. 3(c), $|I_{\parallel}^{M_1^a M_2^a}(t)| = 1$ for all t , indicating that the emission from M_1^a and M_2^a 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 EP 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 EP. Note that the accumulated phase difference between parallel and perpendicular harmonics can be traced back to the TDPs ($\alpha_{\mu}^{\mathbf{k}}$) 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 (see SM). The condition $I_{\parallel}^{M_1 M_2}(t) = -I_{\parallel}^{M_1 M_2}(t + \frac{T}{2})$ ensures purely odd harmonics along the LPD, while $I_{\perp}^{M_1 M_2}(t) = I_{\perp}^{M_1 M_2}(t + \frac{T}{2})$ ensures purely even harmonics perpendicular to the LPD.

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 LPD along $\Gamma - S$, then the eh-PER will be important when $S \neq S_0$. This is illustrated in Fig. 4, where for the $\Gamma - K$ LPD, the excursions of the electron-hole-pairs created at M_1 and M_2 in \mathbf{k} -space, $\boldsymbol{\kappa}(t)$ (teal arrows), are not along the group velocity vector fields $\nabla_{\mathbf{k}} E_n^{\mathbf{k}}$, yielding a nonzero eh-PER. Note that since the double-peaked EP originates in trajectories with similar emission times, they are not expected to be significantly altered by macroscopic effects such as phase matching or intensity averaging [40, 52] [53]. In contrast, when the LPD is $\Gamma - M_1$, the \mathbf{k} -space excursion (purple

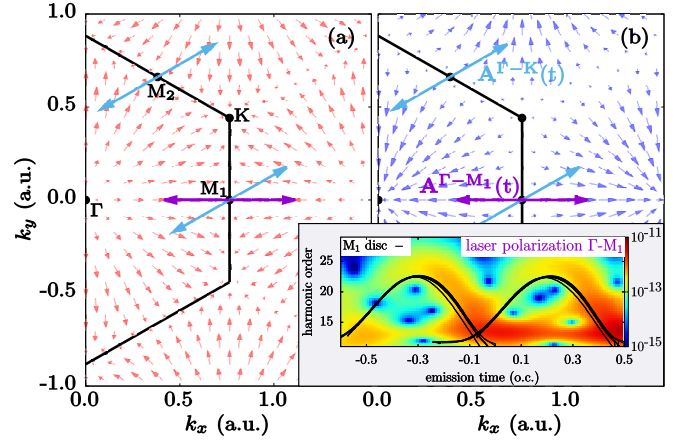


FIG. 4. Relevance of the eh-PER for linearly polarized drivers. The vector fields show the group velocities (without the anomalous term involving the Berry curvature) $\nabla_{\mathbf{k}} E_n^{\mathbf{k}}$ for (a) the hole and (b) the electron. For clarity, the vector-field size in (a) is scaled by 2 compared to (b). The teal (purple) arrows sketch the excursion of the relevant electron-hole pairs in \mathbf{k} -space for a vector potential $\mathbf{A}(t)$ polarized along $\Gamma - K$ ($\Gamma - M_1$). Inset: the EP and the semiclassical predictions, for a driver with $\Gamma - M_1$ polarization.

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 $\Delta \mathbf{r} = \mathbf{0}$. Indeed, this is confirmed by both our quantum and semiclassical calculations in the inset of Fig. 4, where no double peak is observed and all trajectories starting from a M_1 disc in the BZ have approximately the same emission times.

In conclusion, we have uncovered and characterized the effects of imperfect recollisions for HHG in solids. In h-BN, they manifest in the EPs as a double peak structure only for the parallel-polarized harmonics. In the process, we formulated an ERM for HHG in solids that captures the effects of the eh-PER, as well as the implications of the Berry connections and TDPs. We found that the spatial width of the electron and hole wave packets can be almost one order of magnitude larger than the lattice constant, allowing for the imperfect recollisions. This suggests that *spatial* decoherence, caused by imperfections or impurities in the crystal [25, 39, 54–56], may play a role in the rapid *temporal* decoherence frequently included in theoretical models to reproduce experimental spectra [14, 17, 19, 41], and calls for more research into the effect of spatial imperfections on HHG. Furthermore, the agreement between our SBE results and the semiclassical interpretation, supported by our wave packet calculations, favors the picture of the coherent emission of radiation for a (delocalized) electron recolliding with its “own” correlated hole [2, 24], rather than recollision with a “different” uncorrelated one [29].

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 [1, 3, 4, 8, 27, 35, 57, 58] and systems with large Berry curvatures [59], as well as high-order sideband generation [60–63]. The identification of eh-PER and its effect on the harmonic EPs, as well as the formulation of the ERM, provides new insights into the HHG process in solids and could potentially stimulate new experiments, as well as further theoretical developments of a real-space picture of solid-state HHG. 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 important for attosecond metrology in solids [2, 5, 19, 21, 41].

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