The most prominent processes involving the interaction of intense laser fields with matter resulting in nonperturbative, nonlinear dynamics is high-harmonic generation (HHG) [1]. This process, mostly studied in atomic and molecular gases, has resulted in the birth of attosecond science [2], as well as, new ultrafast imaging methods [3,4]. Much less focus had been afforded to corresponding processes in the condensed matter phase. Recent experiments have demonstrated that intense laser-solid interactions offer a wide range of phenomena and applications, including HHG [5–8].

In atoms, the electrons occupy a discrete set bound states; when an electron is ionized it is promoted to the continuum. By contrast, in solids, due to the large number of atoms that make up the lattice, the energy levels become dense and are organized into energy bands. Electrons that are “bound” are contained in the valence band separated from the conduction band by a finite bandgap. When an electron is “ionized” it is promoted to the conduction band leaving behind a hole in the valence band; see Fig. 1. In atoms the hole has a mass that is orders of magnitude greater than the electron and thus its motion can be neglected. In solids, however, electron and hole masses are comparable and thus hole dynamics cannot be ignored.

High-harmonic generation in solids can originate from two channels, an intraband current from nonlinear currents in the individual bands and an interband current due to polarization buildup between the valence and conduction bands; see Fig. 1 for a schematic representation. Theoretically, HHG in solid state systems has been investigated in the single active electron approximation for 1D and 2D band models [9–13]. The focus of the work here is to understand the main driving mechanism of HHG in solids in the underresonant limit. All previous work in this limit has not relied on a full solution of the two-band equations and has exclusively investigated the intraband current. So far the interband current has not been considered.

2. Two-band model for HHG in solids

Our analysis of HHG in solids is based on a two-band tight-binding model coupled to a laser field \( \mathbf{F}(t) = \hat{r}F_0f(t) \); the dipole approximation is used, as the laser wavevector is much smaller
Figure 1. (a) Schematic of the crystal momentum representation of the band structure. (b) Schematic of the energy band structure in space. The intraband current arises from nonlinear currents in the individual bands and the interband current arises from a polarization buildup between the valence and conduction bands.

than the reciprocal lattice vectors. Further, atomic units are used throughout the paper, unless stated otherwise; the laser is characterized by field amplitude $f(t)$. The valence band (holes), the conduction band (electrons) and the bad gap are determined by $E_v = -\Delta E_v$, $E_c = E_g + \Delta E_c$, and $\varepsilon_g = E_c - E_v = E_g + \Delta E_g$, respectively. Intense laser solid interaction is modeled using the two level density matrix equations,

$$\dot{\pi}(K, t) = -\frac{\pi(K, t)}{T_2} - \frac{i}{2} \Omega(K, t) w(K, t) e^{-is(K, t)}$$  \hspace{1cm} (1a)

$$\dot{n}_m(K, t) = i s_m \frac{\Omega^*(K, t)}{2} \pi(K, t) e^{is(K, t)} + c.c.$$  \hspace{1cm} (1b)

where $n_m$ is the probability amplitude of valence ($m = v$) and conduction ($m = c$) band, $w = n_v - n_c$ is the population difference between valence and conduction band. Initially, when all electrons are in the valence band, $n_v = w = 1$; $s_m = -1, 1$ for valence and conduction band, respectively. The crystal momentum $k$ has been transformed into a frame moving with the vector potential $A = -dF/dt$, $K = k - A(t)$. As a result, the first Brillouin zone (BZ) is also shifted to $\overline{BZ} = BZ - A(t)$. Further, $T_2$ accounts for dephasing within the relaxation time approximation, $S(K, t) = \int_{-\infty}^{t} \varepsilon_g(K+A(t'))dt'$ is the classical action, $\Omega(K, t) = 2F(t) \cdot d(K+A(t))$ is the Rabi frequency, and $d(k) = i \int d^3x u_{v,k}^*(x) \nabla_k u_{c,k}(x)$ is the dipole moment. Finally, $\pi$ is connected to the polarization by

$$p(K, t) = d[K+A(t)]\pi(K, t)e^{is(K,t)} + c.c.$$  \hspace{1cm} (2)

The above equations are identical to the two-band semiconductor equations [14] in the single active electron limit. The main difference is the frame of transformation which removes differentiation $\nabla_k$ from the standard two-band equations. This allows for more efficient numerical integration and also allows for generalization of the analytical tools developed for atomic HHG [15] to solids; more detail is given in Ref. [16]. Finally, HHG in solids is determined
by an intraband \( j_{\text{ra}} \) and interband \( j_{\text{er}} \) contribution which are respectively given by [9, 14]

\[
\begin{align*}
    j_{\text{ra}}(t) &= \sum_{m=c,v} \int_{BZ} v_m[K + A(t)] n_m(K, t) \, d^3K \\
    j_{\text{er}}(t) &= \frac{d}{dt} \int_{BZ} p(K, t) \, d^3K,
\end{align*}
\]

(3a)

(3b)

where the band velocity is defined by \( v_m(k) = \nabla_k E_m(k) \). The high-harmonic spectrum is obtained from the Fourier transform (FT) of the total current, \( J_t = j_{\text{ra}} + j_{\text{er}} \), as \( |\text{FT}\{j_t\}|^2 \).

In order to better understand the physical processes driving HHG in solids, it is useful to explore Eqs. (1) by using the Keldysh approximation [17], \( w(t) \approx 1 \) in Eq. (1a). This decouples Eqs. (1) so that they can be formally integrated. In Sec. 3, we will show that the interband term dominates HHG for mid-ir frequencies in ZnO. As such, here we will only focus on the interband term; the intraband term will be considered in future work. Inserting the result into Eq. (3b), we find the interband contribution along laser polarization to be,

\[
    j_{\text{er}}(\omega) = \omega \int_{BZ} \frac{d^3k}{d(k)} \int_{-\infty}^\infty d\tau e^{-i\omega t} \int_{-\infty}^t dt' F(t') d\kappa(t') \times e^{-iS(k, t') - (t-t')/T_2} + \text{c.c.,}
\]

(4)

where \( \kappa(t') = k + A(t') - A(t) \), \( v_m \) and \( d \) are the band velocity and dipole moment along laser polarization. Further, we have transformed the crystal momentum back to the initial frame \( k = K + A(t) \), and \( S(k, t') = \int_{t'}^t \epsilon_g(\kappa_x) dt' \). Note that deriving Eq. (4) directly from the Schrödinger equation would yield the same result except for the dephasing term.

Following the semiclassical method developed for atomic HHG [15], saddle point integration with regard to the three integrals \( d^3k, dt', dt \) in Eq. (4) gives three saddle point conditions,

\[
\begin{align*}
    \nabla_k S &= \Delta x_c - \Delta x_v = 0 \\
    \frac{dS}{dt'} &= \Delta E(k - A(t) + A(t')) = 0 \\
    \frac{dS}{dt} &= \Delta E(k) = \omega.
\end{align*}
\]

(5a)

(5b)

(5c)

The above equations reveal a physical picture for interband HHG. In Eq. (5a), \( \Delta x_m = \int_{t'}^t v_m dt'' = x_m(t') - x_m(t) \) is the distance propagated by the electron/hole in the conduction/valence band \( (m = c, v) \) between time of birth \( t' \) and time of observation \( t \); the velocity is \( v_m = \nabla_k E_m \). This equation reveals a similar process to the acceleration of an electron in the laser field followed by its subsequent re-encounter with its parent ion in atomic HHG. However, in solids the masses of the electron and hole are comparable and thus crystal hole dynamics cannot be neglected. Eq. (5a) then states that, in solids, after having been accelerated and separated by the laser field, HHG takes place when the electron and hole re-encounter one another. Eq. (5b) states that electrons are born with zero momentum at time \( t' \), \( k = A(t) = A(t') \) = 0. The finite bandgap energy results in a complex birth time, which describes tunnel ionization. Finally, Eq. (5c) represents conservation of energy — the electron-hole pair recombines and emits a photon \( \omega \) with energy equal to the bandgap at the momentum of recombination, \( k = A(t) - A(t') \). As a result, the cut-off for interband HHG in a two-band model is limited to the maximum bandgap. Higher harmonics are possible, but require excitation to a higher band.

3. Numerical Results

Our theory is applied using a 3D band structure for ZnO (wurtzite structure). The orientation of the reciprocal lattice is chosen so that \( \hat{x} \parallel \Gamma-M \), \( \hat{y} \parallel \Gamma-K \), and \( \hat{z} \parallel \Gamma-A \) (optical axis).
Figure 2. Harmonic spectrum from interband ($|\mathcal{F}\{j_{\text{IB}}\}|^2$, blue line) and intraband ($|\mathcal{F}\{j_{\text{IN}}\}|^2$, red line) currents for field strength $F_0 = 0.003$; we use a temporal Gaussian envelope with FWHM equal to 10 cycles and cos-carrier with frequency $\omega_0 = 0.014$, corresponding to a laser period $T_0 = 2\pi/\omega_0 = 10.9$ fs; (a), (b), and (c) show dephasing times of $T_2 = \infty$, $T_0/2$, $T_0/4$, respectively. The dashed black vertical lines represent the (minimum) bandgap at the center Brillouin zone.

**3.1. Effect of dephasing time**

Our analysis of HHG begins with a comparison of the two contributions to HHG in Fig. 2. The interband (blue) and intraband (red) currents are shown for dephasing times $T_2 = \infty, T_0/2, T_0/4$ with $T_0$ being the laser oscillation period. Non-perturbative HHG takes place above the dashed line ($N = 9$), indicating the minimum bandgap at the center of the BZ. Both contributions show a plateau in the non-perturbative regime, whereas in experiments the harmonic yield drops with increasing harmonic order. This difference is due to the neglect of propagation effects, in particular reabsorption of harmonic photons which increases with growing harmonic orders. The spectra in Fig. 2 were calculated with peak field strength $F = 0.003$. The calculations were repeated with a range of field strengths used in experiments [5]; all calculations yielded similar results. In the plateau region, interband HHG is always dominant by at least two orders of magnitude.

In Fig. 2(a), both spectra are very noisy and do not show a clear odd harmonic structure. The reason for this noise becomes clear by looking at Figs. 2(b) and (c). The odd harmonic spectrum starts to manifest for dephasing times of the order of a half cycle, see Fig. 2(b).
Figure 3. Interband (circles) and intraband (triangles) contributions to harmonic intensity vs laser wavelength $\lambda_0$ for high-harmonics (HH) 11 – 19 at field strength $F_0 = 0.003$.

For even shorter dephasing times, the signal to background ratio is further improved in 2(c) and a clean harmonic spectrum emerges. This can be understood as follows. In atomic HHG, propagation effects introducing a phase mismatch between the fundamental laser frequency and its second harmonics can also contribute to attenuating second and higher returns [18]. This is not the case in solids, as absorption dominates phase mismatch for above band gap plateau harmonics [13]. This leaves dephasing as the only mechanism for obtaining a clean harmonic spectrum in agreement with experiments.

3.2. Wavelength dependence of the harmonics
Here we describe how the interband and intraband contributions scale with the wavelength of the driving field. Figure 3 shows this scaling for the interband (solid lines) and the intraband (dashed lines) with $T_2 = 2.7$ fs. When the wavelength is reduced, at some point a given harmonic will move beyond the plateau into the cutoff region and the yield will drop. On the other hand, for increasing wavelength a given harmonic order will drop below the band gap and move from the perturbative to the nonperturbative realm. Each harmonic has only been plotted between these two limits — as long as it stays in the plateau region.

The interband contribution drops exponentially with increasing wavelength, which can be attributed to the dephasing term in Eq. (4). By contrast, the wavelength dependence of the intraband yield is more complex and even increases for longer wavelengths. For most of the investigated wavelength range, the interband HHG clearly dominates. Only around wavelengths, where a given harmonic transits from the non-perturbative into the perturbative regime, the two contributions become comparable. The calculations have been repeated for a range of field strengths between $0.002 \leq F_0 \leq 0.006$, which show a very similar picture with small differences in where inter- and intraband current intersect. Our calculations indicate the dominance of interband HHG over the part of the mid-infrared spectrum where non-perturbative...
HHG occurs. Ultimately, the importance of interband versus intraband HHG will have to be settled by experiment. Figure 3 reveals a first suggestion of how this can be done. The opposite gradients of inter- and intraband yield at higher wavelengths present a strong signature for experimentally discerning the two mechanisms.

4. Conclusion
We have presented a 3D, two-band, density matrix analysis for HHG in solids using a single active electron. Numerical results suggest interband HHG, which has been neglected so far, to be the dominant channel. Saddle point integration in the Keldysh/Lewenstein limit reveal the following mechanism responsible for HHG in solids: after the creation of an electron hole pair by tunnel ionization, electrons and holes are accelerated and driven apart by the laser field. When electron and hole re-encounter each other, recombination can occur resulting in the emission of a harmonic photon; the wavevector at the time of recombination determines the energy difference (gap) between the electron-hole pair and therewith the emitted photon energy. By comparison, in atoms an electron is ionized and accelerated by the laser field and ultimately driven back to the core which is stationary due to its weight. Upon recombination it releases its kinetic energy as a high energy photon.

Similar to atomic HHG higher returns and recollisions occur. The influence of higher returns is much stronger in solids than in atoms. In the absence of dephasing, higher returns create a noisy continuum-like structure, which masks the odd harmonic spectrum. Dephasing times of the order of a half cycle are necessary to suppress higher returns and to obtain agreement with the clean odd harmonic structure observed in experiments.

The interband current drops exponentially with increasing laser wavelength. This opens a novel way for measuring dephasing times without the need of ultrashort laser sources. For long wavelengths (> 4μm) the intraband current grows for increasing wavelengths. The different signs of interband and intraband wavelength dependence present a strong signature allowing to experimentally settle the importance of interband versus intraband HHG.

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