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Isolated high-order harmonics pulse from two-color-driven Bloch oscillations in bulk semiconductors

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We theoretically investigate the time-frequency characteristics of high-order harmonic generation (HHG) from Bloch-oscillating electrons in the band structure of a conventional bulk semiconductor driven by a single-optical-cycle two-color IR waveform. Spectrally filtering out the Bloch-HHG cutoff radiation allows the generation of an isolated Bloch-HHG pulse of ∼1.6-fs duration.

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Since the seminal works by Bloch, Zener, and Wannier,[1–3] the feasibility to observe Bloch oscillations (BOs) or equivalently the existence of Wannier-Stark ladders (WSLs) in conventional bulk solids has been the subject of a decades-long heated controversy (an excellent review is given in Ref. 4). As a consequence of the symmetry properties of the crystal structure, an electron under the influence of a static electric field E is accelerated within the electronic band structure according to the acceleration theorem, $\hbar\dot{k} = -eE$ [in one dimension (1D)], and, due to Bragg scattering at the Brillouin zone (BZ) boundaries, performs a periodic ballistic motion in k space accompanied by a real-space oscillation of the electron position, which is known as Bloch oscillation.[1,2] The BO is associated with a Bloch period $T_B = 2\pi/\Omega_B$ and Bloch energy $\hbar\Omega_B = aeE$, which is simply the potential drop over one unit cell with lattice constant a.

The generalization to time-dependent electric fields $E(t)$ is straightforward[5,6]: introducing the instantaneous Bloch frequency via $\hbar\Omega_B(t) = aeE(t)$, the acceleration theorem can be rewritten as $a\dot{k} = -\Omega_B(t)$, and its formal integration yields $k(t) = k_0 + eA(t)/\hbar$ with the initial wave number k_0 and the vector potential $A(t) = -\int_{-\infty}^{t} \mathrm{d}t'\, E(t')$. Thus, the k-space dynamics directly mirrors the vector potential $A(t)$, yet folded into the first BZ via Bragg reflections at the zone boundaries.

The BO picture in k space is equivalent to the WSL picture in real space: in the field-free case, the electron wave functions in a solid are delocalized and the energy spectrum of band states is continuous. When a strong electric field is applied, for which $|aeE|$ is large compared with the width of the band Δ, the electron wave functions become localized and the discrete energy spectrum consists of the WSL, whose eigenenergies are equidistantly spaced by the Bloch energy $\hbar\Omega_B$. Thus, an electronic wave packet is a superposition of these Wannier-Stark states, and the BOs are the quantum beating between these states in time.[5,6] Importantly, Rossi[4] established that the BO and WSL pictures are two totally equivalent rigorously quantum-mechanical representations, which simply correspond to different vector- and scalar-potential gauges, respectively.

BOs and WSLs were first experimentally observed in semiconductor superlattices,[7,8] but meanwhile also in a variety of other artificial systems such as ultracold atoms in standing light waves,[9] Bose-Einstein condensates,[10] degenerate atomic Fermi gases,[11] optical waveguide arrays,[12] optical[13] and ultrasonic[14] superlattices, mesoscopic Josephson junctions,[15] and elastic systems.[16] However, no clear experimental evidence for BOs and WSLs has yet been reported for conventional bulk solids.

Although the formal mathematical problems that were the subject of the above-mentioned BO controversy have been solved,[4] interband Zener tunneling[2] and scattering processes (electron-electron, electron-phonon, impurity scattering, etc.), which we have ignored in our discussion so far, could impede the observation of BOs and prevent the existence of WSLs. Thus, the key question is whether it is possible to make the Bloch period T_B shorter than typical scattering times. In 1998, Rossi arrived at the following answer[4]: "...in the absence of scattering events, BOs exist and, contrary to the early papers by Rabinovitch and Zak,[17] they are not significantly affected by Zener tunneling, both for bulk and superlattices. On the contrary, due to scattering events, BOs are fully suppressed in bulk semiconductors but they still survive in superlattices..." Nevertheless, even though the impossibility to observe BOs in bulk solids has been common sense now for a long time, recent dramatic progress in the generation of intense ultrashort electric fields provides good new reasons for optimism to observe them also in bulk solids.

Already in 2002, we reached a Bloch energy of $\hbar\Omega_B = 3.0\,\text{eV}$ in extreme nonlinear optical experiments on the semiconductor ZnO.[18] In our experiments, 5-fs pulses from a Ti:sapphire laser oscillator were tightly focused to a Gaussian intensity profile with 1-μm radius resulting in a peak electric field of 6 V/nm in a bulk ZnO crystal without observing any damage. As the resulting Bloch period $T_B = 1.4$ fs was just half a cycle of light $T = 2.8$ fs (at photon energy $\hbar\omega_0 = 1.5$ eV), equivalent to a dynamical localization parameter $\Theta_B = \Omega_B/\omega_0 = 2.0$, the electrons might have experienced Bragg reflections within one optical cycle in these experiments.[18] Similar values for $\hbar\Omega_B$ were also achieved in bulk GaAs.[5]

In 2010, Kuehn *et al.* performed time-resolved high-field terahertz experiments to demonstrate ballistic transport of electrons in n-type GaAs across half the BZ (termed a "partial BO," but no Bragg scattering at the BZ boundaries occurred)[19] and to investigate the role of Zener tunneling.[20]

Bloch-oscillating electrons lead to nonperturbative high-order harmonic generation (HHG) of the driving field.[6,21] In 2011, Ghimire *et al.*[22] observed Bloch-HHG up to 25th harmonic order extending to >9.5 eV photon energy, when driving a 500-μm-thick ZnO crystal with 9-cycle-long mid-IR

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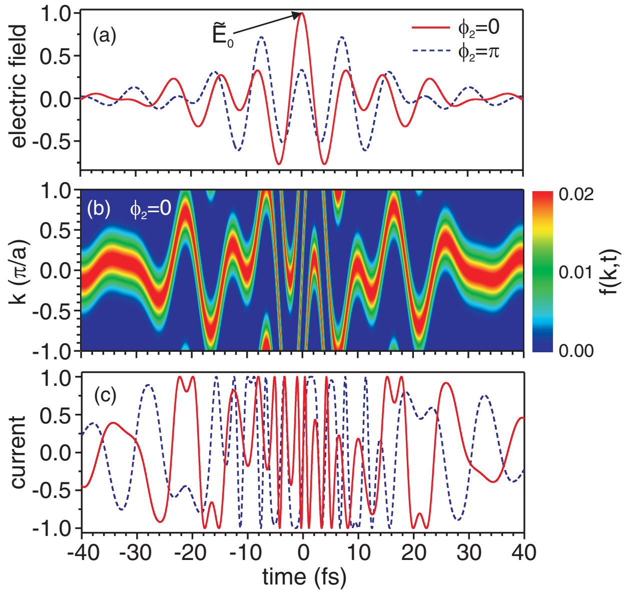

FIG. 1. (Color online) Bloch-oscillating electron wave packet driven by a two-color IR waveform: (a) Electric field $E(t)$ for $\phi_1 = 0$ and two values of ϕ_2. Note the quasi-single-cycle waveform for $\phi_1 = \phi_2 = 0$. (b) Distribution function $f(k,t)$ obtained from the Boltzmann equation (1) for the $\phi_2 = 0$ case [red solid curve in (a)]. (c) Resulting electron current $j(t)$. Parameters are (17 fs, 2.3 μm), ($r_2 = 0.5$, 25 fs, 3.6 μm), $\tilde{E}_0 = 6.2$ V/nm.

pulses (~100-fs, 3.25-μm, 0.38-eV pulses with up to 2.63 μJ energy). Focused field strengths up to 6 V/nm were achieved, exactly the same value as in our earlier ZnO experiments with 0.8-μm pulses.[18] Two observations made in Ref. 22 are particularly noteworthy here: (i) The observation of HHG extending more than 6 eV above the ZnO band gap ($E_{\mathrm{gap}} \sim 3.3$ eV) suggests that these harmonics are generated within a few tens of nm near the output surface of the ZnO crystal. (ii) Interestingly, HHG up to the 25th order was observed for experimental conditions corresponding to $\Theta_B \sim 5$, which directly indicates the cutoff harmonic order.[6,21] We return to this point below.

In this Rapid Communication, motivated especially by the results of Ref. 22, we theoretically explore the opportunities of controlling the time-frequency characteristics of Bloch-HHG by means of incommensurate two-color IR waveforms. In particular, driving the Bloch oscillation by a quasi-single-cycle waveform phase-coherently synthesized from 2.3-μm and 3.6-μm optical fields allows the generation of an isolated HHG pulse of ~1.6-fs duration.

Let us consider a two-color waveform $E(t) = E_0 \times [\mathrm{sech}(t/\tau_1)\cos(\omega_1 t + \phi_1) + r_2\,\mathrm{sech}(t/\tau_2)\cos(\omega_2 t + \phi_2)]$ as driving field with frequencies ω_i and carrier-envelope phases (CEPs) ϕ_i. The parameter E_0 determines the peak electric field $\tilde{E}_0$ of the two-color superposition [occurring at $t = 0$ fs for $\phi_1 = \phi_2 = 0$, see Fig. 1(a)], r_2 is the mixing ratio of field 2. The time constants $\tau_i = t_{\mathrm{FWHM},i}/[2\mathrm{arcosh}(\sqrt{2})]$ are given by the full width at half maximum (FWHM) pulse durations $t_{\mathrm{FWHM},i}$. We emphasize that the pulse parameters used in our calculations are compatible with existing self-CEP-stabilized IR parametric amplifiers.[23]

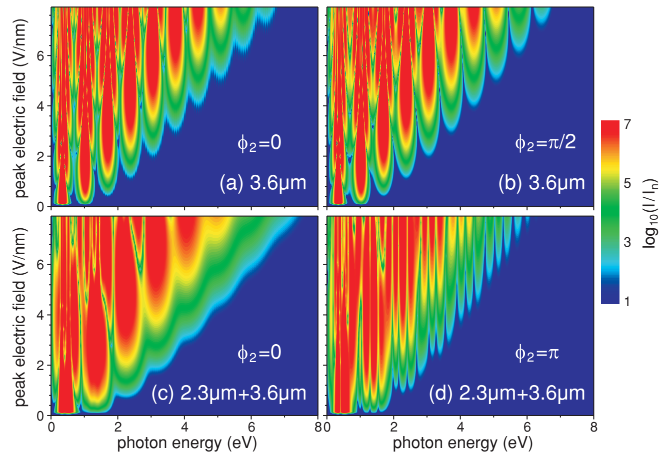

FIG. 2. (Color online) Bloch-HHG spectrum vs peak electric field $\tilde{E}_0$: (a) and (b) Single-color 3.6-μm driver pulse for two values of ϕ_2 as indicated. (c) and (d) Two-color (2.3 μm + 3.6 μm) driver waveform for $\phi_1 = 0$ and ϕ_2 as indicated. Other parameters as in Fig. 1. I_n is a normalization intensity.

This two-color waveform $E(t)$ accelerates a Gaussian Bloch electron wave packet, characterized by a time-dependent distribution function $f(k,t)$ and initially centered at wave number $k_0 = 0$, within a 1D tight-binding (TB) cosine band structure, $\hbar\omega_e(k) = \frac{\hbar^2}{m_e a^2}[1 - \cos(ka)]$ with effective electron mass m_e, according to the Boltzmann equation (scattering processes ignored)

$$\frac{\partial}{\partial t} f(k,t) = -\frac{e}{\hbar} E(t) \frac{\partial}{\partial k} f(k,t). \tag{1}$$

From $f(k,t)$ obtained from numerical solutions of the Boltzmann equation (1) and the electron group velocity $v_g(k) = \frac{d\omega_e}{dk}$, we can compute the generated electron current using $j(t) \propto e \int_{\mathrm{BZ}} dk\, v_g(k) f(k,t)$, where the wave number integration extends over the first BZ.

Figure 1 shows numerical results for a Bloch-oscillating electron wave packet in ZnO ($m_e = 0.24 \times m_0$ with free electron mass m_0; $a = 0.521$ nm for electric field $\vec{E}||\vec{c}$ axis) driven by the two-color waveform $E(t)$. Bragg reflections at the zone boundaries are evident in $f(k,t)$ and lead to strong nonlinearities in the current $j(t)$, which can be controlled within a single optical cycle via the CEP values $\phi_{1,2}$. Note that for $\phi_1 = \phi_2 = 0$ [red curves in Figs. 1(a) and 1(c)], the BO is driven by a quasi-single-cycle waveform and, close

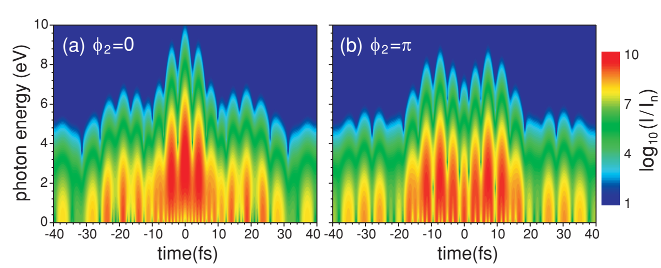

FIG. 3. (Color online) Time-frequency analysis of the Bloch-HHG: the Gabor transforms $G(\omega,t)$ are calculated for the parameters in Fig. 1, $T_G = 0.7$ fs, (a) $\phi_2 = 0$, and (b) $\phi_2 = \pi$.

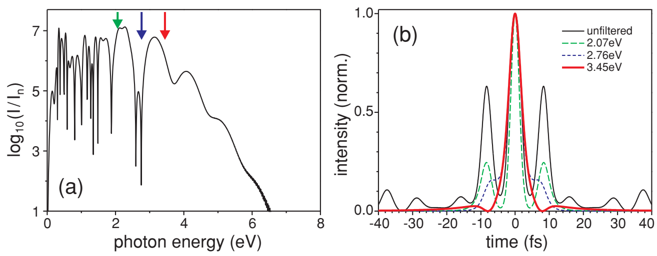

FIG. 4. (Color online) Cutoff filtering of Bloch-HHG: arrows in panel (a) showing the Bloch-HHG spectrum indicate the position of the cutoff filter E_{cutoff} used to obtain the temporal intensity profiles in (b). Parameters as in Fig. 1, $\phi_1 = \phi_2 = 0$.

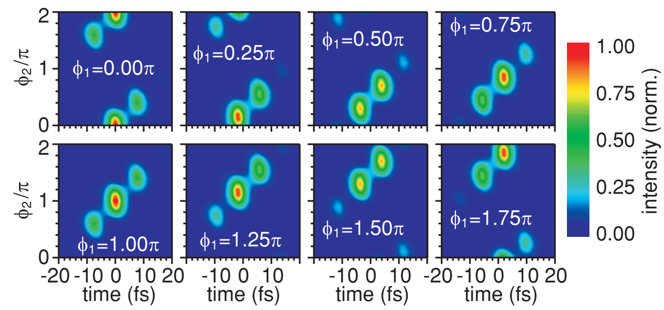

FIG. 6. (Color online) Temporal intensity profile of the Bloch-HHG cutoff radiation vs ϕ_2 for various values of ϕ_1 as indicated. Other parameters as in Fig. 1, $E_{\text{cutoff}} = 3.45$ eV.

to the peak of the electric field at $t = 0$ fs, the resulting current changes from -1 to $+1$ within 640 attoseconds only. Intuitively, detrimental scattering processes are not expected to prevent the BO on such a short time scale (this expectation is supported by the experimental findings in Ref. 22). Moreover, for a maximum experimentally permissible field strength, a single-cycle driver field allows to concentrate the energy into a very short time interval, reducing the risk of damaging the crystal. Since the source term of radiation in the wave equation is given by $\partial j(t)/\partial t$, the spectrum of the emitted radiation can be calculated from $I_{\text{rad}} \propto |\omega j(\omega)|^2$.[6,21]

Recently, several groups employed multicolor driver waveforms for HHG from gases.[24] We point out that HHG described by Corkum's three-step model[25] is distinctly different from the mechanism of Bloch-HHG investigated here: within the three-step model, both the energy of the recolliding electron wave packet and the HHG cutoff scale with $\lambda^2 E^2$, and a multicolor driver waveform permits control over the tunnel ionization (step 1) and coherent steering of the resulting continuum electron wave packet (step 2). In contrast, the Bloch-HHG cutoff is independent of driver wavelength λ and scales linearly with electric field strength E, as confirmed in Fig. 2. For single-color driving and different CEP values ϕ_2 [Figs. 2(a) and 2(b)], only slight differences can be discerned in between different harmonics due to interferences. For two-color driving, either a smooth [Fig. 2(c)] or a strongly modulated Bloch-HHG cutoff region [Fig. 2(d)] is observed depending on the combination of CEP values $\phi_{1,2}$.

To gain deeper insights into the emission dynamics, we perform a time-frequency analysis of the Bloch-HHG shown in Fig. 3 employing the Gabor transform $G(\omega,t) = |\mathcal{F}_{t'}\{[\partial j(t')/\partial t'] \exp[-(t-t')^2/T_G^2]\}|^2$, where $\mathcal{F}_{t'}$ denotes the Fourier transform in t'. Note the inherent tradeoff between time and frequency resolution depending on the time window T_G. As can be seen from the Gabor transforms in Fig. 3, the lower-order harmonics are emitted at several instants during one optical cycle. In contrast, the highest harmonics are only emitted at the extrema of the electric field [compare Fig. 1(a)], which can be controlled via the CEPs $\phi_{1,2}$. Importantly, it does not seem feasible to compress the total bandwidth to an isolated attosecond pulse because of the complicated time-frequency structure of the Bloch-HHG.

These findings suggest that, in analogy to the generation of isolated attosecond XUV pulses via HHG in gases, filtering out the cutoff radiation should permit us to generate an isolated Bloch-HHG pulse as Fig. 4 illustrates. Figure 5 shows the temporal intensity profile of the Bloch-HHG cutoff radiation for two-color driving with $\lambda_2 = 3.6\ \mu$m and variable λ_1. For $\lambda_1 \sim 2.3\ \mu$m, a rather clean isolated pulse is generated for $\phi_2 = 0$ [Fig. 5(a)], while the double-burst emission for $\phi_2 = \pi$ [Fig. 5(b)] is strongly suppressed. The whole dependence of the temporal intensity profile on both CEP values $\phi_{1,2}$ is displayed in Fig. 6. Note in particular that the weights of the peaks close to $t = 0$ fs change as ϕ_1 increases, i.e., it is not simply the relative phase $\Delta\phi = \phi_1 - \phi_2$, but in fact the combination of the CEP values $\phi_{1,2}$ that matters.

The influence of the band structure on the Bloch-HHG can be studied with a modified band structure $\hbar\omega_e(k) = \alpha[1 - \cos(ka)] + \beta[1 - \cos(2ka)]$ introduced in Refs. 26, whose parameters, $\alpha = (-\hbar^4 + 2m_e a^2 \hbar^2 \delta)/(2m_e^2 a^4 \delta)$ and $\beta = \hbar^2/(4m_e a^2) - \alpha/4$, allow one to individually tune the width of the band Δ and the effective mass m_e at the Γ point (i.e., $k = 0$). The results in Fig. 7 reveal that Δ and m_e have a

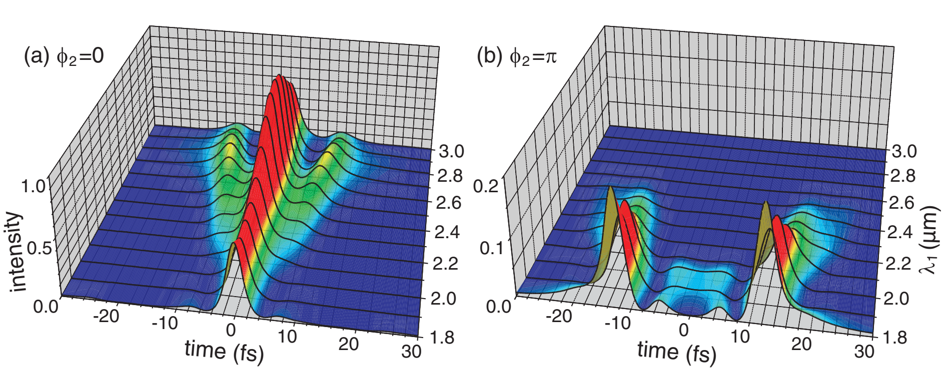

FIG. 5. (Color online) λ_1 dependence of the temporal intensity profile of the Bloch-HHG cutoff radiation for (a) $\phi_2 = 0$ and (b) $\phi_2 = \pi$ (on a rescaled intensity axis). The parameters of field 2 ($r_2 = 0.5$, 25 fs, 3.6 μm) are fixed, field 1 has a fixed 17-fs pulse duration, but its wavelength λ_1 is varied. Other parameters are $\phi_1 = 0$, $\tilde{E}_0 = 6.2$ V/nm, $E_{\text{cutoff}} = 2.76$ eV.

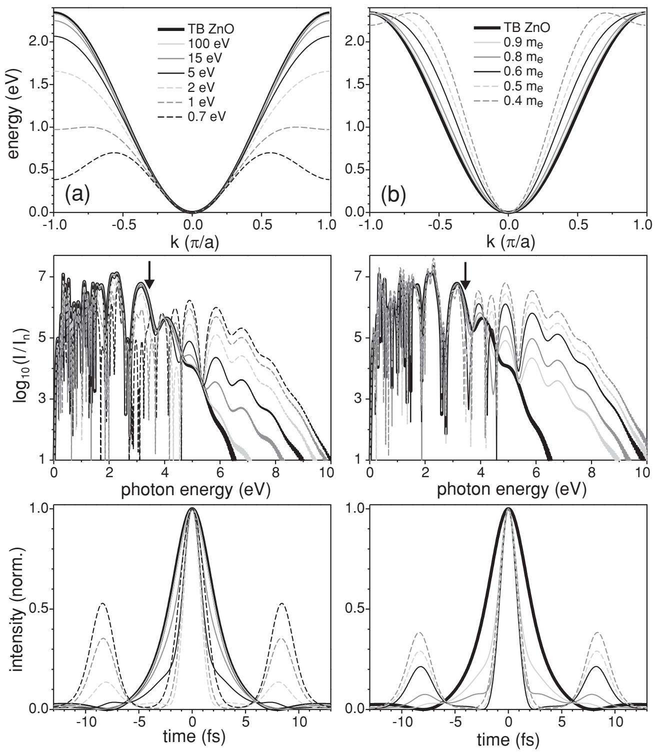

FIG. 7. Influence of the band structure: the columns (a) and (b) show the influence of Δ and m_e, respectively. The results for a TB band with effective mass m_e of ZnO are compared to results for modified band structures for (a) different bandwidth parameters δ and (b) m_e (for fixed Δ) as indicated in top panels. The arrows in the mid panels showing the HHG spectra indicate the cutoff filter ($E_{\text{cutoff}} = 3.45$ eV) used to obtain the temporal intensity profiles in the bottom panels. Parameters as in Fig. 1, $\phi_1 = \phi_2 = 0$.

strong influence on the spectra and time-domain structure of the Bloch-HHG. As argued in Refs. 26, the broadness of HHG emission is mainly determined by the maximum steepness of the band dispersion $\omega_e(k)$ since for a steeper dispersion the intraband acceleration leads to a faster variation of the electron energy via $k(t)$.

In the regime of extreme nonlinear optics,[6] as Golde *et al.*[26] demonstrated within the framework of two-band semiconductor Bloch equations, interband[5,27] and intraband dynamics (i.e., the BOs discussed here) are nontrivially coupled. Their complex interplay leads to a strong enhancement of HHG toward much higher frequencies[26] (this naturally explains the observation of HHG up to 25th order for only $\Theta_B \sim 5$ in Ref. 22) and is also expected to have a profound influence on the time-frequency characteristics of the emitted HHG. Intuitively, it still should be possible to obtain an isolated HHG pulse by spectrally filtering out the cutoff radiation.

In summary, we theoretically investigated the time-frequency characteristics of Bloch-HHG from bulk ZnO driven by two-color IR waveforms. Spectrally filtering out the cutoff radiation allows the generation of an isolated Bloch-HHG pulse of ~1.6-fs FWHM duration. More sophisticated calculations might take into account scattering processes occurring on a few-femtosecond time scale, dressing of the band structure,[28] and study the complex interplay of interband and intraband dynamics.[26]

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