Path integral solution of the Boltzmann-Bloch equation applied to high order harmonic generation in superlattices with asymmetric current flow

Apostolos Apostolakis\textsuperscript{1} and Mauro F. Pereira\textsuperscript{1,2*}

\textsuperscript{1}Department of Condensed Matter Theory, Institute of Physics, Czech Academy of Sciences, Na Slovance 1999/2, 182 21 Prague, Czech Republic
\textsuperscript{2}Department of Physics, Khalifa University of Science and Technology, Abu Dhabi 127788, UAE

(Dated: February 10, 2020)

In this paper we solve the Boltzmann-Bloch equation within a path integral approach, delivering general, non-perturbative solutions of high harmonic generation in semiconductor superlattices with asymmetric current flow. The system is treated non-perturbatively in the illuminating field by employing local boundary conditions which allow the inclusion of asymmetric relaxation rates. The spectroscopic properties of the high harmonic generation are demonstrated by calculations of the nonlinear response in both frequency and time domain. We show that asymmetric currents affect the spontaneous emission and can result in a significant enhancement of even harmonics by tuning the interface quality.

I. INTRODUCTION

The inherent nonlinearities of electronic systems can be exploited for the development of novel compact sources in the terahertz (THz) region [1–5]. The very same nonlinearities and their underlying microscopic origin serve as sensitive means for controlling high harmonic generation (HHG) processes. A notable very recent example is the generation of THz harmonics in a single-layer graphene due to hot Dirac fermionic dynamics under low-electric field conditions [6]. In a parallel effort, advances in strong-field and attosecond physics have paved the way to HHG in bulk crystals operating in a highly nonperturbative regime [7–14]. The first experimental observation of non-perturbative HHG in a bulk crystal was explained on the basis of a simple two-step model in which the nonlinearity stemmed from the anharmonicity of electronic motion in the band combined with multiple Bragg reflections at the zone boundaries [15]. The high frequency (HF) nonlinearities which contribute to harmonic upconversion in bulk semiconductors have been associated to dynamical Bloch oscillations combined with coherent interband polarization processes [7, 9]. The aforementioned models allow the use of tight-binding dispersions [16] to describe the electronic band and therefore the radiation from a nonlinear intraband current. One of the systems that demonstrate a similar highly nonparabolic energy dispersion are man-made semiconductor superlattices (SSLs) [17, 18]. In fact, the possibility of spontaneous frequency multiplication due to the effect of nonparabolicity in a SSL miniband structure was first predicted in the early works of Esaki-Tsu [19] and Romanov [20]. Superlattices are created by alternating layers of two semiconductor materials with similar lattice constants resulting in the formation of a spatial periodic potential. Furthermore, SSLs host rich dynamics in the presence of a driving field, which include the formation of Stark ladders [21], the manifestation of Bragg reflections and Bloch oscillations [22]. From the viewpoint of applications, SSLs have attracted great interest because they allow the development of devices which operate at microwave [23] and far-infrared frequencies [5, 23] suitable for high precision spectroscopic studies and detection of submillimeter waves. In addition, a considerable number of studies have tackled the task of engineering parametric amplifiers [24, 25] and frequency multipliers [26–28] based on superlattice periodic structures. Note that although the first semiconductor superlattice frequency multipliers (SSLM) were developed for the generation of microwave radiation [29], significant progress has been achieved combining high-frequency operation (up to 8.1 THz, ∼50th harmonic) [5, 27] and high conversion efficiency [30] comparable to the performance of Schottky diodes [30, 31].

There are various mechanisms that contribute to the HF nonlinearities of SSL devices. Once this distinction has been clearly made, it is simple to connect the underlying physical mechanisms to the frequency multiplication effects. It was found that spontaneous multiplication takes place in a dc biased tight-binding SSL, when the Bloch-oscillating electron wave packet is driven by the input oscillating field [32, 33]. Moreover, the increase of optical response [32] was due to the frequency modulation of Bloch oscillations [34, 35] which arise in the negative differential conductivity (NDC) region of the current-voltage characteristic, i.e. the current decreases with increasing bias. On the other hand, if a SSL device is in a NDC state, the nonlinearities can be further enhanced by the onset of high-field domains [36, 37] and the related propagation phenomena [38] in a similar way as the electric-field domains in bulk semiconductors [39]. Thus, the ultrafast creation and annihilation of electric domains during the time-period of an oscillating field contributes to harmonic generation processes in SSLs [40]. This type of dynamics has been found to depend on plasma effects [41, 42] induced by the space-charge instabilities and the dielectric relaxation time processes which dictate the exact conditions for the NDC state [42]. The expected THz response from Bloch oscillations in

* mauro.pereira@ku.ac.ae
a miniband SSL, under the influence of a THz electric field, might also deviate due to strong excitonic effects [43–45]. Harvesting the nonlinearities discussed above can potentially lead to more efficient SSLMs or other devices suitable for achieving extremely flexible frequency tuning. Our approach is inspired by very recent theoretical and experimental investigations [28, 46–48] of SSLM behavior, which revealed the development of even harmonics due to imperfections in the superlattice structure. In general, when a adequately strong oscillating field couples energy into the SSLM in the absence of constant bias, only odd harmonics are emitted. However, Ref. [28] showed that symmetry breaking was induced by asymmetric current flow and scattering processes under forward and reverse bias. This approach combined nonequilibrium Green’s function calculations with an Ansatz solution of the Boltzmann equation in the relaxation rate approximation. Furthermore, the asymmetric scattering rates were attributed to the different elastic (interface roughness) scattering rates which have risen from the non-identical qualities of the SSL interfaces. In general, elastic scattering processes can have a significant effect on the electron transport in semiconductor superlattices. The conventional method to study the role of elastic scattering on miniband transport and generation of high-frequency radiation [49, 50] are the one dimensional (1D) SSL Balance equations [49] which can be extended to address two-dimensional and three-dimensional [51, 52] SSL transport and optical properties. They cannot, however, include systematically the different scattering processes under forward and reverse bias. A handful of experiments have been devoted to examine the harmonics of current oscillations [37], transient THz emissions which can be decomposed into \( E_{\text{ac}} \cos(2\pi\nu t) \) and the electron mobility under the influence of isotropic-elastic-scattering time.

In this paper, we elucidate how the effects of asymmetric scattering processes could be used to control the implications of the SL potential on the response of miniband electrons to an oscillating electric field \( E(t) = E_{\text{ac}} \cos(2\pi\nu t) \). Benefiting from the seminal work of Chambers [56] which describes a path-integral approach that is not dependent on any special attributes of the relaxation time, we solve the Boltzmann transport equation to address the asymmetric intraminiband relaxation processes in semiconductor superlattices. Earlier in Refs. [57, 58] the relaxation rate was assumed to depend on the electron velocity allowing to estimate analytically the high-frequency conductivity of an asymmetric superlattice but with resorting to perturbative analysis of the Boltzmann equation. Before proceeding further it is worthwhile first to highlight the main points of this work:

(i) We eliminate the numerical instabilities which originate from the Ansatz solution of Ref. [28, 59] with the Chambers path integral approach.

(ii) We theoretically demonstrate that the multiplication effects can be effectively controlled by special designs of superlattice interfaces (asymmetric elastic scattering). We show that one can gain control over even and odd harmonics by choosing an appropriate asymmetry parameter. These anisotropic effects [28] reflect that typically the interfaces of a host material (A) grown on a different host material (B) are found to be rougher than those of B on A (see Fig. 1), indicating grading or intermixing of the constituent materials between SSL layers [60, 61].

This paper is organized as follows. Section II provides an overview of a semiclassical theory describing the charge transport in SSLs in the presence of asymmetric scattering. In Sec. III, we discuss the nonlinear optical response of miniband electrons in an asymmetric SL structure and we present results of exact numerical simulations describing the spontaneous HHG. Complementary insight is provided next with time-domain calculations. In Appendix A, we revisit in more detail the path-integral expressions implemented in this work.

II. SEMICLASSICAL FORMULATION

Throughout this work we use the standard energy dispersion, \( \epsilon(k_z) = \epsilon^a - 2|T| \cos(k_zd) \), which describes the kinetic energy carried by an electron in the lowest SL miniband [17, 18]. Here \( \epsilon^a \) is the center of the miniband, \(|T|\) is the miniband width, \( k_z \) is the projection of crystal momentum on the \( z \)-axis (axis parallel to the general grown direction) and \( d \) is the superlattice period. Note that in this transport model, the effects of inter-miniband tunneling are neglected. To simulate the temporal distribution function \( f(k,t) \) of the single electron, we employ a semiclassical approach based on the Boltzmann transport equation [18]

\[
\frac{\partial f}{\partial t} + \frac{\mathbf{F}}{\hbar} \frac{\partial f}{\partial \mathbf{k}} = I(f),
\]

where \( \mathbf{F} \) is the force \((-e)E\) corresponding to a time-dependent electric field or a constant electric field applied in the \( z \)-direction of the SSL and \( \mathbf{k} \) is the total momentum which can decomposed into \( k_z \) and the quasi-momentum in the \( x-y \) plane \( k_{\|} = (k_x, k_y) \). The right-hand side term of Eq. (1) represents the collision integral. Instead of using a single relaxation rate approximation model equivalent to \( I(f) = -(f - f_0)\Gamma/\hbar \) with \( f_0(k) \) being the equilibrium fermi distribution, we will resort to two scattering rates \( \Gamma \) to adequately describe the asymmetric relaxation processes. The asymmetric elastic scattering would result to enhanced scattering processes into certain directions. Thus, the kinetic equation can be rewritten in the following form

\[
L^\pm f = \frac{\Gamma^\pm f_0}{\hbar}
\]

where \( L^\pm = 1/\hbar (\mathbf{F}\partial/\partial \mathbf{k} + \hbar\partial/\partial t + \Gamma^\pm) \) are integral operators corresponding to the different relaxation rates (\( \Gamma^+ \) and \( \Gamma^- \)). By using the inverse of the operator, \( L^{-1} \), on the left of Eq. (2) one obtains the generalized Chambers
A. To quantify the effect of asymmetric scattering, we need to consider different starting times [see Eq. (3)]. The SSL sample is biased by an electric field, $E = (0, 0, E_{ac}\cos(2\pi vt))$ parallel to the direction of the $z$-axis.

The path integral

$$L^{-1}\{\phi(k)\} = \int_{-\infty}^{t} dt_{0} \frac{\Gamma(t_{0})\phi[k(t, t_{0})]}{\hbar} \times \exp \left\{ - \int_{t_{0}}^{t} \frac{\Gamma(y)}{\hbar} dy \right\}, \quad (3)$$

where $\phi$ denotes a quantity such as current, displacement, etc. Equation (3) summarizes that an electron which passes through the point $k$ at time $t$, follows different collisionless trajectories which takes it through the points $k(t_{0})$ at times $t_{0} < t$. This compact solution requires the assumption of the following boundary condition

$$\Gamma = \begin{cases} 
\Gamma^{+} & v(t, t_{0}) > 0, \\
\Gamma^{-} & v(t, t_{0}) < 0.
\end{cases} \quad (4)$$

Here $v(t, t_{0})$ represents the time-dependent miniband velocity which reveals the propagation direction of the electron along the sample and therefore indicates the interaction with the high-quality or low-quality interface [see Fig. 1]. For a further discussion of Eqs. (2), (3) see Appendix A. To quantify the effect of asymmetric scattering, we calculate the current density using the approach that was developed above and now takes the form

$$j(t) = \frac{2e}{(2\pi)^{3}} \int d^{3}k f_{0}(k) \int_{-\infty}^{t} dt_{0} \frac{\Gamma(t_{0})v(t, t_{0})}{\hbar\Delta(t, t_{0})} \times \exp \left\{ - \int_{t_{0}}^{t} \frac{\Gamma(y)}{\hbar} dy \right\}, \quad (5)$$

where $k_{z}$ is integrated over the Brillouin zone, the integration limits of the in-plane components $k_{i}$ are $\pm\infty$ and $\Delta(t, t_{0})$ controls the level of current flow asymmetry. The time dependence of the velocity $v(t, t_{0})$ is obtained from the set of the equations

$$\frac{dk_{z}(t, t_{0})}{dt} = \frac{eE(t)}{\hbar}, \quad k_{z}(t, t_{0}) = k_{z0}, \quad (6a)$$

$$v(t, t_{0}) = \frac{2 |T|}{\hbar} \sin(k_{z}d), \quad z(t, t_{0}) = 0. \quad (6b)$$

The peak current $j_{0} = j(E_{c}^{+})$, corresponding to the critical field $E_{c}^{+} = \Gamma^{+}/(ed)$ reads

$$j(E_{c}^{+}) = \frac{2de}{(2\pi)^{3}} \int_{-\pi/d}^{\pi/d} \sin(k_{z}d) dk_{z} \int d^{2}k f_{0}(k). \quad (7)$$

Here again the boundary conditions [Eq. (4)] dictate the different relaxation times $\Gamma(t) = \Gamma^{+}$ or $\Gamma^{-}$, reflecting on the coefficient $\Delta(t, t_{0})=1$ or $\delta$ in Eq. (5). This asymmetry coefficient $\delta = 1^{+}/\Gamma^{-}$ which plays an important role in this work, since it indicates the differences between the interfaces leading to deviation from the perfectly antisymmetric voltage of the Esaki and Tsu model [17]. See Fig. 2. An increase of $\delta$ can be interpreted as a structural variation of the initial SSL structure. In the present work we assume that $\delta \geq 1$ which implies that the flow from left to right will be favored over the flow from right to left. Furthermore, the asymmetry coefficient depends on the elastic and inelastic scattering rates, which are either determined from measured values [37, 63] or nonequilibrium Green’s functions calculations [28, 47]. It is important to notice that similar kinetic formulas to Eq. (5) have been used to treat the different different types of scattering processes in superlattices [64, 65]. However, none of these works have systematically included a tensor analyzing the different relaxation processes which correspond to an asymmetric SSL structure. In this paper, the values of the SSL parameters in Eqs. (5)-(7) are taken from recent experiments and predictive simulations [28, 46]: $d = 6.23$
We see that the asymmetric current flow is dramatically
this paper, which
the asymmetric current flow
a hybrid NEGF-Boltzmann equation approach by em-
As it is one of the main motivations of this paper, which
delivers a clean numerical solution that does not need the

Thus, one can consider a SSL with period \( d \) under an electric field \( E_{dc} + E_{ac} \cos(2\pi vt) \). The time-dependence of the current response is then described by the Fourier basis

\[
j_i(t) = j_{dc}^i + \sum_{l=1}^{\infty} J_{l}^{i,\cos}(\alpha_jt) \cos(2\pi \nu t) + j_{l}^{i,\sin}(\alpha_jt) \sin(2\pi \nu t),
\]

where the dc current \( j_{dc}^i \) is given by Eq. (9) and the Fourier components \( \{j_{l}^{i,\cos}(\alpha), j_{l}^{i,\sin}(\alpha)\} \) describe the \( l \)-th harmonic generation. Here the harmonic-conversion properties of the SSLM critically depend upon the strength of the nonlinear response through the parameter \( \alpha = eE_{dc}d/(h\nu) \). The terms \( J_n(\alpha) \) in Eqs. (9)–(11) denote the Bessel functions of the first kind and order \( n \). It can also be seen from Eq. (9) that the voltage current (VI) characteristics in the presence of irradiation \( E(t) \) is given by a sum of shifted Esaki-Tsu characteristics

\[
E_{dc}(U) = j_0(2U/\Gamma)/[1 + (U/\Gamma)^2]
\]

where \( j_0 \) is the peak current corresponding to the critical electrical field \( E_c = h/(e\nu) \). This might lead to a photon-assisted tunneling phenomenon that has been experimentally observed [18, 67]. Moreover, note that the term \( U = eE_{dc}d + nh\omega \) designates an effective potential difference instead of the plain potential drop per period due to the dc bias. The function \( K(U) = 2j_0/[1 + (U/\Gamma)^2] \) is connected to \( j_{dc} \) through Kramers-Kronig relations. The intensity of the emitted radiation from the SSL structure is determined by the Poynting vector, which is proportional to the harmonic current term [28]

\[
I_{\nu}^2(t) = 2 \langle j(t) \cos(2\pi \nu t) \rangle^2 + \langle j(t) \sin(2\pi \nu t) \rangle^2,
\]

where the integration \( \langle \cdot \rangle \) signifies time-averaging over time interval of infinite time in the general case. Nevertheless, considering that the current response is induced merely by a monochromatic field \( \nu \), it is sufficient to average only over the time-period \( T = 1/\nu \). It is thus sufficient for our studies to look at the resulting average

\[
\langle I_2 \rangle^2 = 2 \langle j(t) \cos(2\pi \nu t) \rangle^2 + \langle j(t) \sin(2\pi \nu t) \rangle^2.
\]
At this point we should give a brief recap of the analytical rigorous numerical approach; the ansatz is implemented flow \[28, 46–48\], to compare its predictions to those of our ansatz previously used to describe asymmetric current BOHF oscillations. Harmonics of the oscillating field is related only to the SSL remains uniform. As a result, the gain at some leads to a single-electron state and the electric field within the frequency field considered to be acting on the superlattice operates in the NDC region might result to the forma-
tion of high electric field domains which act as additional characteristic. As mentioned above, a superlattice which to an active state equivalent to the NDC region of the VI from the ac-field in order to bring temporarily the SSL α > α

\[
\alpha = \frac{\alpha}{\alpha} + \frac{\Gamma}{\Gamma} - \frac{U}{U} > 0,
\]

Here \( \alpha \) is the asymmetry coefficient \( \delta \) used in the time domain calculations depicted in Fig. 6(c) and Fig. 7(d) respectively. The color bar is normalized to the peak current \( j_0 \).

\[
I_l^{(\nu)} = (j_l^{(\nu, \cos)})^2 + (j_l^{(\nu, \cos)})^2,
\]

In order to investigate spontaneous frequency multiplication effects. Both even and odd harmonics are present in a biased SSL due to symmetry breaking. However, in this paper we focus on symmetry breaking due structural effects leading to asymmetric current flow. Thus in all numerical results for harmonic generation, there is no static electric field, i.e. \( U = eE_{dc,d} = 0 \) and the Bragg reflections from mini-zone boundaries are not associated to a specific oscillation period \( v_B = eE_{dc,d}/h \). On the other hand, the Bragg scattering is manifested as frequency modulation of electron oscillations during a cycle of the oscillating field. Oscillations of this type are known as Bloch oscillations in a harmonic field (BOHF). Due to intraminiband relaxation an electron performs high quality BOHF when \( \alpha > \alpha_c \). Here \( \alpha_c = U_c/(h\nu) \) and \( U_c = \Gamma \) is the energy required from the ac-field in order to bring temporarily the SSL to an active state equivalent to the NDC region of the VI characteristic. As mentioned above, a superlattice which operates in the NDC region might result to the formation of high electric field domains which act as additional linearities. We must underline that in this work the high-frequency field considered to be acting on the superlattice leads to a single-electron state and the electric field within the SSL remains uniform. As a result, the gain at some harmonics of the oscillating field is related only to the nonlinearity of the voltage-current characteristic and the BOHF oscillations.

At this point we should give a brief recap of the analytical ansatz previously used to describe asymmetric current flow \[28, 46–48\], to compare its predictions to those of our rigorous numerical approach; The ansatz is implemented by replacing \( j_0 \) in Eqs. (8)-(11) by

\[
\left\{ \begin{array}{ll}
    j_0 & U > 0, \\
    \frac{j_0}{\gamma} & U < 0
\end{array} \right. \quad \Gamma = \left\{ \begin{array}{ll}
    \Gamma^+ & U > 0, \\
    \Gamma^- & U < 0.
\end{array} \right.
\]

where the potential energy \( U \) is equal to integer number of photon quanta \( n\hbar\nu \) and the asymmetry coefficient is \( \delta = j_0/J_0 = \Gamma^+ / \Gamma^- \). This approach predicted the development of even harmonics in good agreement with experiments \[28, 46\]. Revisiting the latter approach allows the straight comparison with the solution developed in Sec II. The basic idea is to vary the asymmetry coefficient \( \delta \) which in both approaches is defined as the ratio of the different relaxation rates \( \Gamma^\pm \) and then examine the effects on even and odd order harmonics. Figure 3 depicts the second harmonic (left-handed panels) and third harmonic output (right-handed panels) as a function of the \( \alpha \) parameter for different values of the asymmetry coefficient \( \delta \). The dependencies \( |I_l(\nu)|^2 \) (a) were calculated using Eqs. (8)–(13) and Eqs. (5), (12) in Fig. 3(a) and Fig. 3(b), respectively. Both approaches yield similar results for the second harmonic in a wide range of \( \alpha \). In particular, we highlight that asymmetric relaxation times are an unconventional mechanism for frequency doubling in SSLs. As \( \delta \) increases, the frequency doubling effects become more pronounced and, eventually, give rise to stronger optical response almost up to 0.6 %. We note that the Ansatz solution may, however, contribute to nonphysical numerical instabilities by revealing intense second harmonic generation even at small amplitudes of the oscillating field. Therefore, the numerical solution offers a reliable way to treat the scattering induced asymmetries in the current flow. Now we turn our attention to the third harmonic output in the presence of asymmetric

\[
\begin{array}{c}
\end{array}
\]

\[
\begin{array}{c}
\end{array}
\]
current flow which is quite different from the behavior of the second harmonic output. Moreover, increasing \( \delta \) suppresses it, implying a redistribution of spectral components in favor of even harmonics as shown in Fig. 3(d). One can see that the maximum output of the third harmonic might be potentially reduced from 20 to 15%. In this case the different solutions appear to be more consistent with each other. However, as shown in the inset of Fig. 3(d) the Ansatz solution can lead to numerical instabilities comparable with the maximum output of the second harmonic. Once we established that the approach developed in this work affords the significant variation of the asymmetry coefficient, we can have an in-depth look into the HHG processes.

Further insight on how asymmetric effects can result in a significant gain at some even harmonic frequencies and suppression at some other odd-order harmonics, is given by the color maps in Fig. 4. It shows the calculated values \( |\mathcal{I}_2(\nu)|^2 \) as a function of \( \alpha \) and \( \delta \). The black area indicates values \( (\alpha, \delta) \) for which \( \mathcal{I}_2 \) exhibits small or negligible harmonic response. The colored areas unfold distinct islands of significant harmonic response. For example, Fig. 4(a) reveals significant enhancement of \( I_2 \) for \( 1.2 < \delta < 1.4 \) and \( 10 \lesssim \alpha \lesssim 70 \). On the other hand, in the same region Fig. 4(d) demonstrates a weak third harmonic response of the irradiated superlattice. The corresponding island of enhanced \( I_3 \) is shifted to significantly larger \( \alpha \) values. The magnitude of \( I_3 \) increases approximately from \( \delta = 1.2 \) to \( \delta = 1 \) and thus obtaining a maximum for a SSL structure with perfectly symmetric interfaces. The width of the islands changes significantly for the higher-order even harmonics as shown in Figs. 4(b) and 4(c). However, although the width of the colored islands is increased, the strength of the harmonic content is reduced, by an order of magnitude for \( I_6 \) [Fig. 4(c)] in comparison with \( I_2 \) [Fig. 4(a)]. The colored areas of the higher-odd harmonics are notably suppressed in the regions where higher-even harmonics are being developed. Therefore, in order to achieve easily detectable odd harmonics the SSL should operate deep inside the NDC region. At this point it is important to highlight that the higher the harmonic order, the larger the input power must be. However, note that arbitrarily increasing the input power is not a solution for high nonlinear output, in contrast with materials described by conventional susceptibilities. There is a complex combination of asymmetry and power values leading to maximum HHG generation. For example, Fig. 5 demonstrates the output of higher even-order harmonics (beyond the 2nd harmonic) which drastically drops when the input power is significantly larger. The SSL device after excitation by a strong GHz input signal can generate measurable 8th harmonic up to \( \sim 0.02\% \). The magnitude of the emitted power in units \( \mu W \) is related to harmonic term \( \mathcal{I}_l \) as \( P_l(\nu) = T \mathcal{I}_l^{2}(\nu) \) where the coefficient \( T = A \mu_0 c L^2/(8\pi n_r) \) obtained from the time-averaged Poynting vector by neglecting the waveguide effects. Here \( \mu_0 \) is the permeability and \( c \) is the speed of light by considering both of them in free-space.

For typical mesa area \( A = (10 \times 10) \mu m^2 \), effective path length through the crystal \( L = 121.4 \) nm and refractive index \( n_r = \sqrt{13} \) (GaAs), one can obtain \( T \simeq 77 \mu W \). Now it is straightforward to calculate the emitted power corresponding to Figs. (3-5). As a consequence, for a value \( \alpha \approx 34 \) close to but below the \( \alpha_c \), the emitted power can reach the values \( P_2 = 0.4 \mu W \) and \( P_4(\nu) = 0.01 \mu W \) at room temperature for the second and fourth harmonic respectively. These magnitudes indicate that significant gain can appear at second and fourth-order harmonics in the absence of electric domains which might affect the HHG processes when \( \alpha > \alpha_c \).

Next, we complement the steady-state analysis with calculations of the time-dependent nonlinear response of the miniband electrons. Our time-dependent solution [see Eq. 5] can provide further insight in the frequency-conversion of the input signal related to the asymmetric scattering processes. Figure 6 depicts the oscillating field, the nonlinear current oscillations, the second harmonic component and the third harmonic component which occur in the presence of asymmetric scattering rates. The oscillating field \( E(t) \) [see Fig. 7(a)] causes a time-dependent electron drift with a time dependent current \( j(t) \) which contains different harmonic components due to the enhanced nonlinear response as shown in Fig. 6(b). In a perfectly symmetric structure, the irradiation of the superlattice with input radiation leads only to odd-order multiplication and therefore the second harmonic signal \( j_2(t) = j(t) \cos(4\pi \nu t) \) [dashed curve in Fig. 6(c)] averaged over time is \( < j_2 >_t \approx 0 \). On the contrary, for a higher asymmetry parameter \( \delta \) (arrowed), the time realization of \( j_2(t) \) demonstrates oscillations whose amplitude is highly asymmetric. In this case, the the first peak (1) becomes sufficiently smaller than peak (2) resulting in \( < j_2 >_t \) different than zero as is evident from Fig. 6(c). The third harmonic component in the current is due to the BOHF which stem from the anharmonic motion of the electron within the miniband. Every half-period \( T/2 \), \( j_3 \) contributes a phase of an opposite sign with respect to the temporal evolution of the electric field [see Fig. 6(a), (d)]. With increasing asymmetry coefficient \( \delta \), the
amplitude of the arrowed peak is reduced, which leads to suppression of the third harmonic component $<j_3>$. For a electric field with sufficiently larger amplitude but with the same oscillating frequency, the current response becomes evidently more anharmonic [see Fig. 7(b)]. This has important implications for both second and third-order harmonics and serious consequences in the case of increasing the asymmetry parameter $\delta$. On the one hand, the increase of the asymmetry between the two relaxation rates results in more pronounced differences between the oscillations amplitudes (1), (2) of the second harmonic $j_2(t)$ and their adjacent peaks [Fig. 7(c)]. Consequently, the second harmonic is suppressed for a larger $E_{ac}$ but still enhanced for a different $\delta$. On the other hand, the $\alpha$ parameter being larger than $\alpha_c$ would induce higher quality BOHF and therefore larger third harmonic components [Fig. 7(d)]. We note though that a larger asymmetry will reduce the emission of $j_3$ due to the strong suppression of the closely neighboring peaks to the main one.

In summary, the Boltzmann-Bloch equation within a path integral approach is used to deliver general, non-perturbative solutions of High Harmonic Generation in semiconductor superlattices. This approach allows us to investigate details of the generation processes in both spectral and time domains. The non-approximative nature of our approach eliminates numerical errors for small harmonics which could cast doubt upon the origin of harmonic generation. Thus, our study conclusively demonstrates striking features of High Harmonic Generation when asymmetric relaxation processes are taken into account in superlattice structures. While these effects are relatively small on the odd harmonic generation, significant features appear at even harmonics leading to measurable effects in the GHz-THz range.

**IV. CONCLUSIONS**

FIG. 6. (Color online) Nonlinear response of miniband electrons by considering asymmetric scattering processes. (a) The normalized electric field $|E(t)/E_{ac}|$ which causes the time dependent drift. (b) The time-dependent current $j(t)$ [see Eq. (5)] is depicted over two cycles of the input field $E(t)$. (c) The second-harmonic $j_2(t)$ and (d) the third-harmonic current oscillations $j_3(t)$ calculated for different values of the asymmetry parameter $\delta = 1, 1.05, 1.2, 1.4$. The labels (1) and (2) denote relevant relative minimum and maximum points. In all cases, the value of the parameter $\alpha \simeq 27$ corresponds to an electric field with amplitude $E_{ac} = 0.75 E_c$ and oscillating frequency $\nu = 141$ GHz. The arrow marks increasing asymmetry.

FIG. 7. (Color online) Nonlinear response of miniband electrons by considering asymmetric scattering processes. (a) The normalized electric field $|E(t)/E_{ac}|$ which causes the time dependent drift. (b) The time-dependent current $j(t)$ [see Eq. (5)] is depicted over two cycles of the input field $E(t)$. (c) The second-harmonic $j_2(t)$ and (d) the third-harmonic current oscillations $j_3(t)$ calculated for different values of the asymmetry parameter $\delta = 1, 1.05, 1.2, 1.4$. The labels (1) and (2) denote relevant relative minimum and maximum points. In all cases, the value of the parameter $\alpha \simeq 86$ corresponds to an electric field with amplitude $E_{ac} = 2.4 E_c$ and oscillating frequency $\nu = 141$ GHz. The arrow marks increasing asymmetry.
the parametric processes can affect the harmonic generation [70] or the Bloch gain [69] profile in the presence of asymmetric current flow. Moreover, our approach has a great potential for analyzing the effects of asymmetric scattering processes on the intensity of harmonics by means of externally applied voltages. Finally, it’s worth considering further the deviations from a completely anti-symmetric current-voltage characteristic and analyze the nonlinear response of SSL excited by a Gaussian optical pulse [45, 53].

V. ACKNOWLEDGMENTS

The authors acknowledge support by the Czech Science Foundation (GAČR) through grant No. 19-03765 and the EU H2020-Europe’s resilience to crises and disasters program (H2020–grant agreement no. 832876, AQUA3S). Access to computing and storage facilities owned by parties and projects contributing to the National Grid Infrastructure MetaCentrum provided under the programme “Projects of Large Research, Development, and Innovations Infrastructures” (CESNET LM2015042) is greatly appreciated.

Appendix A: Path-integral expressions

In this section, we revisit path-integral expressions describing a solution to transport problems and having as a starting point the Boltzmann equation. The general formalism has been applied to describe transport in semiconducting devices [71], parametric amplification [24] and Bloch gain [72] in spatially homogeneous SSLs. This method allows us to deliver a general numerical solution for the influence of asymmetric relaxation effects on miniband transport model and frequency multiplication processes in superlattices in the presence of an oscillating electric field, eliminating the need for the approximative Ansatz used in Refs. [28, 46–48]. The electron distribution function \( f(k, t) \) satisfies the spatially homogeneous Boltzmann equation

\[
\frac{\partial f}{\partial t} = - \frac{F}{\hbar} \frac{\partial f}{\partial k} + \int dk' [f(k') W(k', k) - f(k) W(k, k')].
\]

The second term of the right-hand side of Eq. (A1) represents the rate of change of \( f \) due to collisions, which is characterized conventionally by the transition probability \( W(k', k) dk \) per unit time that an electron will be scattered out of a state \( k \) into a volume element \( dk \) and the rate \( W(k, k') dk \) per unit time that an electron with wave vector \( k \) will scatter to a state whose vector lies between \( k' \) and \( dk' \). We can rearrange Eq. (A1) as

\[
\frac{1}{\hbar} \left( \frac{F}{\hbar} \frac{\partial}{\partial k} + \frac{\partial}{\partial t} + \Gamma \right) f = L f
\]

(A2)

Thus, if a single and isotropic (same for all states \( k \)) relaxation rate is assumed then \( L f = \Gamma f_0 / \hbar \) and Eq. (A2) has an exact formal solution in the form a path-integral

\[
f(k, t) = \int_{-\infty}^{t} dt_0 \frac{\Gamma(t_0) f_0(k(t_0))}{\hbar} + \int dk' f(k') W(k', k(t)) \exp \left\{ - \int_{t_0}^{t} \frac{\Gamma(y)}{\hbar} dy \right\},
\]

(A3)

We would like to localize the asymmetry of the electron scattering function due to interface roughness to well-defined regions of the SSL. Therefore, we assume that \( W(k_z', k_z(t_0)) = W_0 \) if \( k_z' \) and \( k_z \) both lie in within a region of the miniband for which \( v_z(k_z', k_z(t_0)) > 0 \), otherwise \( W(k_z', k_z(t_0)) = 0 \). Accordingly, the operator \( L \) is generalized into

\[
L^+ = \frac{1}{\hbar} \left( \frac{F}{\hbar} \frac{\partial}{\partial k} + \hbar \frac{\partial}{\partial t} + \Gamma^+ \right) \quad \text{(A4a)}
\]

\[
L^- = \frac{1}{\hbar} \left( \frac{F}{\hbar} \frac{\partial}{\partial k} + \hbar \frac{\partial}{\partial t} + \Gamma^- \right) \quad \text{(A4b)}
\]

and

\[
\Gamma^+ / \Gamma^- = 1 + (W_0 / \Gamma^-) \int_{\text{region}^+} f(k') dk'
\]

(A5)

indicating the existence of two scattering rates due to differences in interface roughness depending on the sequence of the layers. Here for simplicity we designate the region in \( k \)-space as \( \text{region}^+ \) corresponding to the high-quality interface of the SSL. Equations (A2) and (A4a), (A4b) can be combined into the single integro-differential Eq. (2). The latter equation may be solved to obtain the current with asymmetric relaxation processes. The resulting expression is described by

\[
j(t) = \frac{2e}{(2\pi)^3} \int dk f_0(k) \int_{-\infty}^{t} dt_0 \frac{\Gamma(t_0) v(t, t_0)}{\hbar \Delta(t, t_0)} \times \exp \left\{ - \int_{t_0}^{t} \frac{\Gamma(y)}{\hbar} dy \right\}.
\]

(A6)

Note that the static current \( j_{dc} \) is obtained by taking \( \Delta(0, t) \) in Eq. (A6).

We should comment here that our approach is qualitatively different from the balance equations approach developed in [73] and discussed further in Refs. [49, 50]. This 1D model assumed that the distribution function can be decomposed into its symmetric \( f_s = \{ f(\mid k , t) + f(\mid -k , t) \}/2 \) and anti-symmetric \( f_a = \{ f(\mid k , t) - f(\mid -k , t) \}/2 \) parts. The basic idea is that \( f_a \) in the presence of inelastic scattering processes \( \Gamma_{in} \) is allowed to relax to equilibrium distribution function \( f_0 \). On the other hand, \( f_a \) couples the motion only in the \( z \)-direction.
This model predicts effectively the suppression of peak current density with the increase of $\Gamma_{el}$. It cannot, however, treat in its present form the asymmetric relaxation rates and their effects on harmonic generation in the presence of a time-dependent electric field, in contrast to our more general approach.

[1] SS Dhillon, MS Vitiello, EH Linfield, AG Davies, Matthias C Hoffmann, John Booske, Claudio Paoloni, M Gensch, Peter Weightman, GP Williams, et al., “The 2017 terahertz science and technology roadmap,” Journal of Physics D: Applied Physics 50, 043001 (2017).
[2] Masayoshi Tonouchi, “Cutting-edge terahertz technology,” Nature photonics 1, 97 (2007).
[3] Fran¸cois Blanchard, Gargi Sharma, Luca Razzari, Xavier Ropagnol, Heidi-Christina Bandulet, Fran¸cois Vidal, Roberto Morandotti, Jean-Claude Kieffer, Tsumeuyuki Ozaki, Henry Tiedje, et al., “Generation of intense terahertz radiation via optical methods,” IEEE Journal of Selected Topics in Quantum Electronics 17, 5–16 (2010).
[4] Zhigang Chen and Roberto Morandotti, “Nonlinear photonics and novel optical phenomena,” Vol. 170 (Springer, 2012).
[5] Vladimir Vaks, “High-precise spectrometry of the terahertz frequency range: the methods, approaches and applications,” Journal of Infrared, Millimeter, and Terahertz Waves 33, 43–53 (2012).
[6] Hassan A Hafez, Sergey Kovalev, Jan-Christoph Deinert, Zoltán Mics, Bertram Green, Nilesh Awari, Min Chen, Semyon Germanskiy, Ulf Lehner, Jochen Teichert, et al., “Extremely efficient terahertz high-harmonic generation in graphene by hot dirac fermions,” Nature 561, 507 (2018).
[7] Olaf Schubert, Matthias Hohenleutner, Fabian Langer, Benedikt Urbanek, C Lange, U Huttner, D Golde, T Meier, M Kira, Stephan W Koch, et al., “Sub-cycle control of terahertz high-harmonic generation by dynamical bloch oscillations,” Nature Photonics 8, 119 (2014).
[8] F. Langer, M. Hohenleutner, C.P. Schmid, C. Poellmann, P. Nagler, T. Korn, C. Schiller, 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 (2016).
[9] Matthias Hohenleutner, Fabian Langer, Olaf Schubert, Matthias Knorr, U Huttner, SW Koch, M Kira, and Rupert Huber, “Real-time observation of interfering crystal electrons in high-harmonic generation,” Nature 523, 572 (2015).
[10] F. Langer, Hohenleutner, M.U. Huttner, Koch, M. S.W., Kira, and R. Huber, “Symmetry-controlled temporal structure of high-harmonic carrier fields from a bulk crystal,” Nature Photon. 11, 227 (2017).
[11] F. Langer, Hohenleutner, M.U. Huttner, Koch, M. S.W., Kira, and R. Huber, “Symmetry-controlled temporal structure of high-harmonic carrier fields from a bulk crystal,” Nature 557, 76 (2018).
[12] Markus Drescher, Michael Hentschel, Reinhard Kienberger, Gabriel Tempea, Christian Spielmann, Georg A Reider, Paul B Corkum, and Ferenc Krausz, “X-ray pulses approaching the attosecond frontier,” Science 291, 1923–1927 (2001).
[13] Olga Smirnova, Yann Mairesse, Serguei Patchkovskii, Nirit Dudovich, David Villeneuve, Paul Corkum, and Misha Yu Ivanov, “High harmonic interferometry of multi-electron dynamics in molecules,” Nature 460, 972 (2009).
[14] Ferenc Krausz and Misha Ivanov, “Attosecond physics,” Reviews of Modern Physics 81, 163 (2009).
[15] Shambhu Ghimire, Anthony D DiChiara, Emily Sistrunk, Pierre Agostini, Louis F DiMauro, and David A Reis, “Observation of high-order harmonic generation in a bulk crystal,” Nature physics 7, 138 (2011).
[16] D Golde, T Meier, and Stephan W Koch, “High harmonics generated in semiconductor nanostructures by the coupled dynamics of optical inter-and intraband excitations,” Physical Review B 77, 075330 (2008).
[17] Leo Esaki and Ray Tsu, “Superlattice and negative differential conductivity in semiconductors,” IBM Journal of Research and Development 14, 61–65 (1970).
[18] Andreas Wacker, “Semiconductor superlattices: a model system for nonlinear transport,” Physics Reports 357, 1–111 (2002).
[19] R Tsu and L Esaki, “Nonlinear optical response of conduction electrons in a superlattice,” Applied Physics Letters 19, 246–248 (1971).
[20] IUA Romanov, “Nonlinear effects in periodic semiconductor structures(frequency multiplication due to non-parabolicity of dispersion law in semiconductor structure subbands, noting electromagnetic signal transformation),” Optika i Spektroskopiia 33, 917–920 (1972).
[21] EE Mendez, F Agullo-Rueda, and JM Hong, “Stark localization in gaas-gaals superlattices under an electric field,” Physical review letters 60, 2426 (1988).
[22] Christian Waschke, Hartmut G Roskos, Ralf Schwedler, Karl Leo, Heinrich Kurz, and Klaus Köhler, “Coherent submillimeter-wave emission from bloch oscillations in a semiconductor superlattice,” Physical review letters 70, 3319 (1993).
[23] P Khosropanah, A Baryshev, W Zhang, W Jellema, JN Hovenier, JR Gao, TM Klapwijk, DG Pavleve, BS Williams, S Kumar, et al., “Phase locking of a 2.7 thz quantum cascade laser to a microwave reference,” Optics letters 34, 2958–2960 (2009).
[24] Karl Friedrich Renk, Benjamín Ingo Stahl, Andreas Rogl, T Janzen, DG Pavleve, Yu I Koshurinov, V Ustinov, and A Zhukov, “Subterahertz superlattice parametric oscillator,” Physical review letters 95, 126801 (2005).
[25] KF Renk, A Rogl, and BI Stahl, “Semiconductor-superlattice parametric oscillator for generation of subterahertz and terahertz waves,” Journal of luminescence 125, 252–258 (2007).
[26] Florian Klappenberger, Karl Friedrich Renk, P Renk, Bernhard Rieder, Yu I Koshurinov, DG Pavleve, V Ustinov, A Zhukov, N Maleev, and A Vasilyev, “Semiconductor–superlattice frequency multiplier for gen-
[55] H Sakaki, T Noda, K Hirakawa, M Tanaka, and T Matsusue, “Interface roughness scattering in GaAs/AlAs quantum wells,” Applied physics letters 51, 1934–1936 (1987).

[56] RG Chambers, “The kinetic formulation of conduction problems,” Proceedings of the Physical Society. Section A 65, 458 (1952).

[57] GM Shmelev, II Maglevanny, and AS Bulygin, “Current-voltage characteristic of asymmetric superlattice,” Physica C: Superconductivity 292, 73–78 (1997).

[58] GM Shmelev, NA Soina, and II Maglevannyi, “High-frequency conductivity of an asymmetric superlattice,” Physics of the Solid State 40, 1574–1576 (1998).

[59] The theory developed in [28] leads to results similar to those obtained in the present work qualitatively and numerically. However, the enhanced asymmetries cause numerical instabilities for a small parameter $\alpha = eEd/\hbar \omega$.

[60] R ´ aM Feenstra, D ´ aA Collins, DZ-Y Ting, M ´ aW Wang, and T ´ aC McGill, “Interface roughness and asymmetry in InAs/GaSb superlattices studied by scanning tunneling microscopy,” Physical review letters 72, 2749 (1994).

[61] Y Tokura, T Saku, S Tarucha, and Y Horikoshi, “Anisotropic roughness scattering at a heterostructure interface,” Physical Review B 46, 15558 (1992).

[62] N.W. Ashcroft and N.D. Mermin, Solid State Physics (Saunders College, Philadelphia, 1976).

[63] A Patane`, D Sherwood, L Eaves, TM Fromhold, M Henini, PC Main, and G Hill, “Tailoring the electronic properties of GaAs/AlAs superlattices by InAs layer insertions,” Applied physics letters 81, 661–663 (2002).

[64] TM Fromhold, A Patane`, S Bujkiewicz, PB Wilkinson, D Fowler, D Sherwood, SP Stapleton, AA Krokhin, L Eaves, M Henini, et al., “Chaotic electron diffusion through stochastic webs enhances current flow in superlattices,” Nature 428, 726 (2004).

[65] MT Greenaway, AG Balanov, E Schöll, and TM Fromhold, “Controlling and enhancing terahertz collective electron dynamics in superlattices by chaos-assisted miniband transport,” Physical Review B 80, 205318 (2009).

[66] William H Press, Saul A Teukolsky, William T Vetterling, and Brian P Flannery, Numerical recipes in Fortran 77: the art of scientific computing, Vol. 2 (Cambridge university press Cambridge, 1992).

[67] Andreas Wacker, Antti-Pekka Jauho, Stefan Zeuner, and S. James Allen, “Sequential tunneling in doped superlattices: Fingerprints of impurity bands and photon-assisted tunneling,” Phys. Rev. B 56, 13268–13278 (1997).

[68] Timo Hyart, Natalia V Alexeeva, Ahti Leppänen, and Kirill N Alekseev, “Terahertz parametric gain in semiconductor superlattices in the absence of electric domains,” Applied physics letters 89, 132105 (2006).

[69] Yu A Romanov and Yu Yu Romanova, “Self-oscillations in semiconductor superlattices,” Journal of Experimental and Theoretical Physics 91, 1033–1045 (2000).

[70] Timo Hyart, Alexey V Shorokhov, and Kirill N Alekseev, “Theory of parametric amplification in superlattices,” Physical review letters 98, 220404 (2007).

[71] Herbert Budd, “Path variable formulation of the hot carrier problem,” Physical Review 158, 798 (1967).

[72] Timo Hyart, Jussi Mattas, and Kirill N Alekseev, “Model of the influence of an external magnetic field on the gain of terahertz radiation from semiconductor superlattices,” Physical review letters 103, 117401 (2009).

[73] A Ignatov, “Self-induced transparency in semiconductors with superlattices,” Soviet Physics-Solid State 17, 2216–2217 (1975).