Coherent control of electric currents in superlattices and molecular wires: Effect of relaxation

Kirill A. Pronin[1,2] and Andre D. Bandrauk[1]
[1]*Universite de Sherbrooke, Sherbrooke, Quebec, Canada J1K 2R1*
[2]*Institute of Biochemical Physics, Russian Academy of Sciences, Moscow 119 991, Russia*
(Received 17 May 2003; revised manuscript received 17 February 2004; published 21 May 2004)

We consider a *d*-dimensional conductor (a superlattice) within the independent-electron one-band approach taking into account relaxation processes. The nonperturbative nonlinear dc electric current in response to a sum of coherent time-periodic electric fields with frequencies ω and 2ω and a phase shift φ is studied in (a) the quantum coherent ''dynamic'' regime and (b) the ''kinetic'' regime under the influence of scattering. For slow relaxation the first one takes place at short time and the second at long time. We demonstrate that coherent control of dc electric currents is possible in both cases, though the particular conditions and manifestations are drastically different. We obtain a detailed picture of coherent control through intraband dynamics, and discuss the role of scattering and the time evolution of the response.

DOI: 10.1103/PhysRevB.69.195308 PACS number(s): 73.21.−b, 78.67.−n, 72.10.−d, 42.65.Ky

I. INTRODUCTION

The usual way of producing a current in a semiconductor is by applying an electric field, which changes the velocity of the carriers and their concentration in semiconducting layers. A novel and promising alternative to a dc bias is coherent control—the quantum effect of controlling the magnitude and direction of the electric current through phase relationships of the applied coherent ac fields. Typically a laser field ω and its generated second harmonic 2ω with some phase shift φ are used—without any dc field component. Changing the phase φ and the amplitudes of the components $\mathbf{E}_1$, $\mathbf{E}_2$, one controls the magnitude and even the polarity of the produced dc current. Many aspects of the effect have been studied lately for semiconductors, superlattices, optical lattices, and molecular and quantum wires—both theoretically and experimentally.[1–30]

Phase-coherent control of the dc current, generated in shallow-level doped semiconductors by multifrequency laser excitation has been considered in Ref. 17. That was an extension to solid-state devices of the previous work by the authors on controlling gas phase reaction products.[18] Coherent control through the carrier photoexcitation from the ground state in the quantum well up to the continuum has been used for the interpretation of experimental data in Ref. 19. Calculations of phase-controlled interband transitions in bulk semiconductors have been performed in Ref. 20. Rectification of the harmonic-mixing field in a single-band tight-binding system with quantum dissipation (coupling to a thermal bath of harmonic oscillators) has been studied in Ref. 23. The space-time symmetry aspects of the directed diffusion and dc current in ac field, both in the classical and quantum framework, have been investigated in Ref. 25. Phase control of the emitted THz field in the calculation for the dynamical Franz-Keldysh effect has been demonstrated numerically for the two-band model of a quantum well with the account of excitonic effects.[26]

Experimentally phase-controlled currents in quantum-well superlattices have been measured in Ref. 19 with the use of mid-infrared radiation and its second harmonic. Coherently controlled directional photocurrents due to interband one- and two-photon excitation in bulk semiconductor GaAs have been studied experimentally in Ref. 21. The most interesting systems for the observation of coherent-control effects in the solid state include semiconductor dots and superlattices,[1–9] optical lattices,[9–12] quantum wires,[6,9] and molecular wires.[13–15]

Typically coherent control is considered to determine the current at the stage of photoexcitation of the carrier from the bound donor state, Ref. 17, or from the ground state in the well to the continuum,[19] or across the band gap.[20] In our approach, on the contrary, we consider the carriers to be already present either intrinsically or due to some unspecified mechanism, and focus on intraband propagation. One of the primary goals of the paper is to demonstrate that coherent control is possible through intraband evolution within simple constant-time relaxation approach, both in the quantum relaxation-free case and in the case of slow scattering. The comparison of the latter two cases reveals the role of coherence and relaxation in phase control and demonstrates the nontrivial time dependence of the response.

Closely related to the problem of coherent control is the effect of dynamic localization, discovered in. Ref. 29. In the present paper we use our results from the study of dynamic localization,[30,31] extending Ref. 29, and relate this to asymmetric ionization of molecules with two-color laser excitation.[16,32,33]

II. MODEL AND GENERAL SOLUTION

We consider a *d*-dimensional crystalline conductor (a semiconductor/optical superlattice, molecular/quantum wire) within an independent-electron one-band approach. The lattice structure is of cubic type [linear in 1D (one dimensional), square in 2D, cubic in 3D]. The overlap between all sites is taken into account, and the transfer integrals might be anisotropic, so that the electron dispersion $H^{(0)}(\mathbf{k})$ is of rather general type. The system is exposed to a two-mode time-periodic (period T, basic frequency ω) space-homogeneous electric field

$$\mathbf{E}(t)=\mathbf{E}_1\cos(\omega t)+\mathbf{E}_2\cos(2\omega t+\varphi), \qquad (1)$$

which may be strong compared to the second energy-type parameter $\hbar\omega$. In the Wannier basis the quantum Hamiltonian reads

$$H(t)=\sum H^{(0)}_{\mathbf{n},\mathbf{n}'}|\mathbf{n}\rangle\langle\mathbf{n}'|+e\mathbf{E}(t)\sum \mathbf{n}|\mathbf{n}\rangle\langle\mathbf{n}|. \qquad (2)$$

Here $\mathbf{n}$, $-\infty<n_i<\infty$, enumerates the lattice sites. The sign of the electron charge is incorporated in the equations here and below, so that e is its modulus. Below we consider explicitly the electrons. The generalization to the case of holes is straightforward—by the change of the overlap integral $H^{(0)}_{\mathbf{n},\mathbf{n}'}$ in the dispersion relation and of the sign of the charge e. However, we do not address the simultaneous presence of both types of carriers, as that would need the account of correlation and excitonic effects beyond the adopted independent-electron approximation.

We are interested in the study of the dc response to the ac electric field, Eq. (1). The former is characterized by the electric current $\mathbf{j}(t)=-e(d/dt)\Delta\langle\mathbf{R}(t)\rangle$, where $\Delta\langle\mathbf{R}(t)\rangle$ is the average displacement of the electrons in the field, summed over the band filling $\Delta\langle\mathbf{R}(t)\rangle=\Sigma\mathbf{n}[\rho_{\mathbf{n},\mathbf{n}}(t)-\rho_{\mathbf{n},\mathbf{n}}(0)]$ (ρ—the density matrix). The angular brackets here and below denote quantum averaging in the dynamic relaxation-free case, while in the kinetic regime it is averaging with the quantum density operator over scattering and fluctuations. Double angular brackets denote additional time average over the period of the field.

The Schrödinger equation of the system without relaxation (2) can be solved exactly to give (see, for example, Refs. 29 and 30)

$$\Delta\langle\mathbf{R}(t)\rangle=\int d\mathbf{k}\frac{\rho_{\mathbf{k},\mathbf{k}}(0)}{V_{BZ}}\int_0^t dt'\,\mathbf{v}_{\mathbf{k}(t')},$$

$$\mathbf{v}_{\mathbf{k}(t)}=\frac{1}{\hbar}\nabla_{\mathbf{k}}H^{(0)}[\mathbf{k}(t)],\quad \mathbf{k}(t)=\mathbf{k}+\frac{e}{\hbar c}\mathbf{A}(t). \qquad (3)$$

Here

$$\mathbf{A}(t)=-c\int_0^t dt\,\mathbf{E}(t)$$
$$=-\frac{c}{\omega}\mathbf{E}_1\sin(\omega t)-\frac{c}{2\omega}\mathbf{E}_2[\sin(2\omega t+\varphi)-\sin\varphi] \qquad (4)$$

is the vector potential and $\mathbf{A}(0)=0$. The scalar potential is identically zero. $H^{(0)}(\mathbf{k})$ is the electron dispersion in the absence of the field and $\rho_{\mathbf{k},\mathbf{k}}(0)$ is the initial density matrix, $\rho_{\mathbf{k},\mathbf{k}'}=\Sigma\rho_{\mathbf{n},\mathbf{n}'}\exp(-i\mathbf{k}\cdot\mathbf{n}+i\mathbf{k}'\cdot\mathbf{n}')$. V_{BZ} is the volume of the Brillouin zone. We take the intersite distance a as length unit throughout.

Scattering by phonons is introduced by dephasing the wave function and inducing relaxation to the thermal equilibrium distribution $f(\varepsilon(\mathbf{k}))$.[29,34] We do not specify the particular form of the equilibrium distribution function—the solution and subsequent discussion is good for a generic $f(\varepsilon(\mathbf{k}))$. The equation for the density matrix $\rho(t)$ is of the form

$$i\hbar\frac{\partial}{\partial t}\rho_{\mathbf{n},\mathbf{n}'}(t)=[H^{(0)},\rho(t)]_{\mathbf{n},\mathbf{n}'}+e\mathbf{E}(t)(\mathbf{n}-\mathbf{n}')\rho_{\mathbf{n},\mathbf{n}'}(t)$$
$$-i\alpha\hbar(1-\delta_{\mathbf{n},\mathbf{n}'})\rho_{\mathbf{n},\mathbf{n}'}(t)$$
$$-i\alpha\hbar\,\delta_{\mathbf{n},\mathbf{n}'}\left[\rho_{\mathbf{n},\mathbf{n}'}(t)-\frac{N_e}{N_s}f(\varepsilon(\mathbf{k}))\right], \qquad (5)$$

where α is the relaxation rate. The density matrix has the norm $\Sigma\rho_{\mathbf{n},\mathbf{n}}(t)=N_e$, where N_e is the total number of electrons and N_s is the number of sites in the lattice. The equilibrium distribution function $f(\varepsilon(\mathbf{k}))$ is normalized to 1 and is symmetric in $\pm\mathbf{k}$. The solution of Eq. (5) is (see, for example, Refs. 29 and 30)

$$\mathbf{j}(t)=-e\int d\mathbf{k}\frac{\rho_{\mathbf{k},\mathbf{k}}(0)}{V_{BZ}}e^{-\alpha t}\mathbf{v}_{\mathbf{k}+(e/\hbar c)\mathbf{A}(t)}$$
$$-e\alpha N_e\int d\mathbf{k}\,f(\varepsilon(\mathbf{k}))e^{-\alpha t}\int_0^t dt'$$
$$\times e^{\alpha t'}\mathbf{v}_{\mathbf{k}+(e/\hbar c)\mathbf{A}(t)-(e/\hbar c)\mathbf{A}(t')}. \qquad (6)$$

Despite the different appearance, Eq. (6) is in fact equivalent to Eq. (4.5) of Ref. 29 (generalized to the case of arbitrary lattice structure, long-distance overlap, and arbitrary initial conditions). However, the form of Eq. (6) presented here is more convenient for analytical analysis and makes transparent the relation to conventional formulas of solid-state theory.

The first term in Eq. (6) characterizes the exponential decay of the initial coherent oscillations, damped by relaxation. At low scattering $\alpha T\ll 1$ (T is the period of the field), and short time $\alpha t\ll 1$ the first term governs the evolution—it is independent of α, while the second one is small with αT. This limit in the main term is equivalent to the coherent relaxation-free case (3) when α is strictly zero and formula (3) is good in the entire time domain. Now back in the kinetic regime $\alpha\neq 0$, the second term in Eq. (6) describes the standard kinetic evolution plus the transition process. It becomes dominant at long time $\alpha t\gg 1$ when the first term fades. The first relaxation-free regime we call dynamic (or short-time), the second one kinetic (long time). Note that it is impossible to go back to the short-time dynamic regime from the long-time kinetic one even in the limit of vanishing scattering. Indeed, the transition to the long-time limit at small (but finite) damping implies that the electron during that time has suffered many scatterings and thus has thermalized. This precludes us in fact from going back to the dynamic (relaxation-free) regime of electron evolution by subsequent transition to vanishing scattering.

III. COHERENT CONTROL AND RELAXATION

Let us analyze the possibility of controlling the dc electron response by the applied coherent ac fields and the effect

of relaxation upon electron propagation/coherent control on the basis of Eqs. (3) and (6).

First, we exclude from the very beginning the case of strong scattering $\alpha T \gg 1$ which corresponds to the conventional kinetic regime with coherent effects vanishing—this is not of interest to us.

For the study of coherence control the case of slow scattering ($\alpha T \ll 1$) is of major interest, and it will be addressed in the rest of the paper. At short time ($\alpha t \ll 1$) and for low scattering the purely coherent (dynamic) results with no relaxation, Eqs. (2) and (3), studied previously,[29–31] are reproduced. This is the first interesting case which we will address in the following section, aimed at coherent control of dc currents in the dynamic regime. The second important case deals with the kinetic regime at long time $\alpha t \gg 1$ and low scattering $\alpha T \ll 1$, which we will address in the following section. In this second case the electron after many scatterings has thermalized, but the probability of a scattering event during one period of the applied ac field is still low, so that coherent effects in electron evolution are still of primary importance. The comparison of these two cases will reveal the effect of slow relaxation upon coherent control of electric currents.

To relate the present consideration to our previous study of dynamic localization[30] we note that the applied field, Eq. (1), has no dc component, which positions the present case as the ''periodic'' regime of the cited references. In the periodic case the quantum evolution of the electronic system $\mathbf{k}(t)=\mathbf{k}+(e/\hbar c)\mathbf{A}(t)$ during the period of the external field drives it back to the same states in $\mathbf{k}$ space it occupied before. However, in position space the electron may shift during that time. This nonzero on average dc drift in ac fields (or in other words, zero-harmonic generation) is the subject of interest for coherent control of electric currents.

A. Dynamic (or short-time) regime

The expression of the dc current in the dynamic relaxation-free regime follows readily from Eq. (3). Besides, we use the electron dispersion in a d-dimensional cubic-type lattice for the velocity in the band to give

$$\mathbf{v}_{\mathbf{k}} = -\frac{2}{\hbar}\sum_{\mathbf{n}>0} H^{(0)}_{\mathbf{0},\mathbf{n}}\mathbf{n}\sin(\mathbf{k}\cdot\mathbf{n}). \tag{7}$$

We also assume the initial band filling symmetrical in $\pm\mathbf{k}$, $\rho_{\mathbf{k},\mathbf{k}}(0)=\rho_{-\mathbf{k},-\mathbf{k}}(0)$, no currents in the initial state in the absence of the field. Then the expression of the average dc current is

$$\langle\langle \mathbf{j}(t)\rangle\rangle = -\frac{e}{\pi\hbar}\sum_{\mathbf{n}} H^{(0)}_{\mathbf{0},\mathbf{n}}\mathbf{n} \times \int d\mathbf{k}\frac{\rho_{\mathbf{k},\mathbf{k}}(0)}{V_{BZ}}\cos(\mathbf{k}\cdot\mathbf{n})I_{dyn}(\varepsilon_1,\varepsilon_2;\varphi), \tag{8}$$

where $\varepsilon_1=\mathbf{n}\cdot e\mathbf{E}_1/\hbar\omega$, $\varepsilon_2=\mathbf{n}\cdot e\mathbf{E}_2/2\hbar\omega$ are the dimensionless field amplitudes, normalized by field frequencies, and

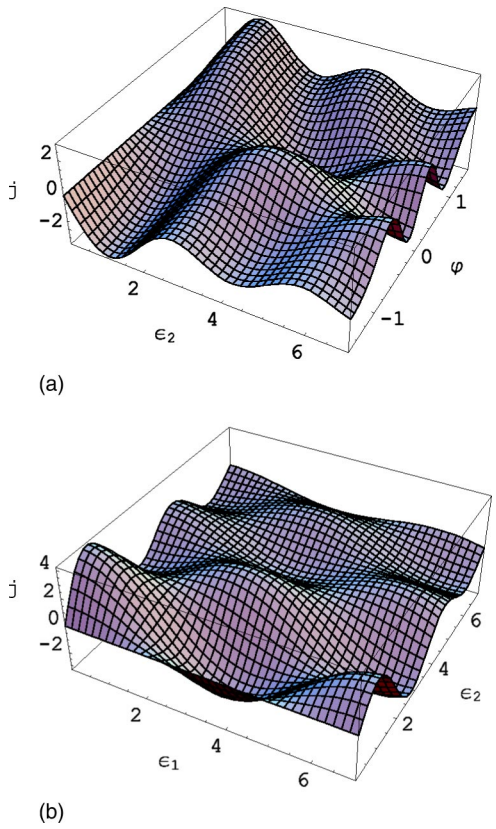

FIG. 1. The dc electric current $\langle\langle j\rangle\rangle$ (arbitrary units) in the coherent dynamic regime (a) as function of the phase shift φ in the range $[-\pi/2,\pi/2]$ and of the dimensionless field amplitude ε_2 with $\varepsilon_1=1$ fixed, and (b) as function of ε_1, ε_2 for phase shift $\varphi=\pi/2$.

$$I_{dyn}(\varepsilon_1,\varepsilon_2;\varphi)=\int_0^{2\pi} d\tau \sin[\varepsilon_1\sin\tau+\varepsilon_2\sin(2\tau+\varphi) - \varepsilon_2\sin\varphi]. \tag{9}$$

We note that the same expression (8) can be obtained from the kinetic case below, Eq. (14), in the limit $\alpha t \ll 1$, $\alpha T \ll 1$.

To avoid unnecessary complications let us adopt the tight-binding approximation when summation over $\mathbf{n}$ corresponds to nearest neighbors only.

Let us study the ε and φ dependencies of the current. The symmetry properties in φ:

$$\langle\langle \mathbf{j}(t)\rangle\rangle\big|_{-\varphi} = -\langle\langle \mathbf{j}(t)\rangle\rangle\big|_{\varphi}, \quad \langle\langle \mathbf{j}(t)\rangle\rangle\big|_{\pi-\varphi} = \langle\langle \mathbf{j}(t)\rangle\rangle\big|_{\varphi} \tag{10}$$

follow readily from Eq. (9) and reduce the study to the range $0\leqslant\varphi<\pi/2$. For $\varphi=0$ the current is zero identically. Clearly, increasing the phase shift φ from $-\pi/2$ to $\pi/2$ one obtains the change of the electric current from some positive (negative) value to some negative (positive) one, depending on the values of ε_1 and ε_2, see Fig. 1(a). Thus, in the dynamic regime both the directionality and the magnitude of the current can be changed for φ in the range $-\pi/2<\varphi<\pi/2$.

The $\varepsilon_1,\varepsilon_2$ dependencies of the current $\langle\langle j\rangle\rangle$ in the dynamic regime show current reversals, depicted in Fig. 1(b) for $\varphi=\pi/2$. Obviously, changing either one or both of the parameters $\varepsilon_1,\varepsilon_2$ along a suitable trajectory one can go from positive values of the current through zero to negative values, or vice versa. For example, from the maximal value $-I_{dyn}=4.050\ldots$ at $\varepsilon_1=0$, $\varepsilon_2=1.031\ldots$ to the minimum $-I_{dyn}=-3.185\ldots$ at $\varepsilon_1=3.634\ldots$, $\varepsilon_2=0.9631\ldots$, $\varphi=\pi/2$. As it can be seen from Fig. 1(b), the decrease of one (or both) component(s) of the applied field may lead to counterintuitive increase of the resulting dc current, and vice versa.

Explicit analytic expansions can be obtained for small values of ε. For example, for $\varepsilon_2\ll 1$ and arbitrary ε_1 one gets

$$I_{dyn}(\varepsilon_1,\varepsilon_2;\varphi)=2\pi\varepsilon_2\sin\varphi[J_2(\varepsilon_1)-J_0(\varepsilon_1)]-\cdots. \tag{11}$$

For ε_2 strictly zero the current $\langle\langle j\rangle\rangle$ is zero identically. The growth rate for small ε_2 is linear, with the slope dependent on the value of ε_1 and sign and value of the phase shift φ.

More interesting is the case $\varepsilon_1\ll 1$ with ε_2 arbitrary:

$$\begin{aligned} I_{dyn}(\varepsilon_1,\varepsilon_2;\varphi)=&-2\pi\sin[\varepsilon_2\sin\varphi]J_0(\varepsilon_2)\\ &+\frac{\pi}{2}\varepsilon_1^2\{\sin[\varepsilon_2\sin\varphi]J_0(\varepsilon_2)\\ &+\sin\varphi\cos[\varepsilon_2\sin\varphi]J_1(\varepsilon_2)\}-\cdots. \end{aligned} \tag{12}$$

Here the current is nonzero even for $\varepsilon_1=0$, first term in Eq. (12). Thus, with the change of the magnitude of the only component ε_2 with $\varepsilon_1=0$, we obtain the dc current whose value changes from positive to negative values and back. The difference with the previous case, Eq. (11), is in the phase shift φ for the field ε_2. This phase shift cannot be compensated by the shift of the time origin, even in the absence of the other field ε_1. In fact, φ determines the phase at which the applied field starts at $t=0$, and this effect does not die away even at long time due to the absence of relaxation in the dynamic regime. Note that the solutions, Eqs. (3) and (6), imply that the vector potential satisfies $\mathbf{A}(t=0)=0$. Thus in the simplest case $\varepsilon_1=0$, $\varepsilon_2\neq 0$ the coherent control of the sign and magnitude of the current is due rather to the initial-field-value effect. However, if both fields ε_1, ε_2 are present, their coherent interference comes into play as well.

The second term in the low ε_1 expansion (12) is not linear, but quadratic in ε_1.

Both expansions, Eqs. (11) and (12), reproduce correctly the limits $I_{dyn}(\varepsilon_1,\varepsilon_2;\varphi\to 0)\to 0$, and $I_{dyn}(\varepsilon_1\to 0,\varepsilon_2\to 0;\varphi)\to 0$. The low $\varepsilon_1,\varepsilon_2$ expansion

$$I_{dyn}(\varepsilon_1\ll 1,\varepsilon_2\ll 1;\varphi)=-2\pi\varepsilon_2\sin\varphi+\cdots \tag{13}$$

signifies that the lowest-order contribution to the dc current is the linear one in ε_2. The form of the last expansion is quite transparent: for low quasimomentum transfer $(e/\hbar c)A(t)\ll 1$ (low field) the average of the velocity, Eq. (3), is proportional to the time-average $e/\hbar c<A(t)>=\varepsilon_2\sin\varphi$, Eq. (4).

Equation (13) can be compared to the drift velocity in two-color photoionization of simple molecules:[32,33] $v_d(t_0)=(E_1/\omega)\sin(\omega t_0)+(E_2/2\omega)\sin(2\omega t_0+\varphi)$, where ωt_0 is the phase around the field maximum (minimum) where ionization occurs mainly. In the present case the latter reduces to the initial time $t_0=0$. Obviously, the corresponding drift velocity $v_d(t_0=0)$ agrees with the expansion (13).

Next, we pass over to the kinetic regime with relaxation.

B. Kinetic (or long-time) regime

In the periodic regime the integral over time in the general kinetic solution, Eq. (6), for the electric current can be represented as a sum of integrals over periods of the applied field, and the corresponding geometric series can be easily summed up.[30,31] The dc current at time $t=mT$ (where m is integer) is given by

$$\begin{aligned} \langle\langle\mathbf{j}(t=mT)\rangle\rangle=&-e\int d\mathbf{k}\frac{\rho_{\mathbf{k},\mathbf{k}}(0)}{V_{BZ}}\frac{e^{-\alpha mT}}{T}\\ &\times\int_0^T d(\Delta t)e^{-\alpha\Delta t}\mathbf{v}_{\mathbf{k}+(e/\hbar c)\mathbf{A}(\Delta t)}\\ &-\frac{e\alpha N_e}{T}\frac{1-e^{-\alpha(m+1)T}}{1-e^{-\alpha T}}\int d\mathbf{k}f(\varepsilon(\mathbf{k}))\\ &\times\int_0^T d(\Delta t)e^{-\alpha\Delta t}\int_0^T d(\Delta t')\\ &\times e^{\alpha\Delta t'}\mathbf{v}_{\mathbf{k}+(e/\hbar c)A(\Delta t)-(e/\hbar c)\mathbf{A}(\Delta t')}. \end{aligned} \tag{14}$$

Note that in the absence of scattering $\alpha=0$ the first term of Eq. (14) reproduces the dynamic result of the preceding section. However, as soon as we pass to the long-time limit $\alpha t\gg 1$ in Eq. (14) the first term fades and there is no way back to the dynamic regime any more.

As discussed above, of major interest to us is the long-time $\alpha t\gg 1$, low-scattering $\alpha T\ll 1$ regime, when the electrons have thermalized, but coherence effects are still of primary importance, as scattering during the period of the ac field is a rare occasion. Then the leading term of the dc current is

$$\begin{aligned} \langle\langle\mathbf{j}\rangle\rangle=&-\frac{eN_e}{T^2}\int d\mathbf{k}f(\varepsilon(\mathbf{k}))\int_0^T d(\Delta t)\\ &\times\int_0^T d(\Delta t')\mathbf{v}_{\mathbf{k}+(e/\hbar c)\mathbf{A}(\Delta t)-(e/\hbar c)\mathbf{A}(\Delta t')}. \end{aligned} \tag{15}$$

However, it can be shown easily that on a cubic-type lattice with symmetric $\pm\mathbf{k}$ distribution function $f(\varepsilon(\mathbf{k}))=f(\varepsilon(-\mathbf{k}))$ the expression (15) is identically zero due to symmetry relations for any periodic field $\mathbf{E}(t)$ without dc component.

Next-order terms in the expansion in αT are nonzero:

$$\langle\langle \mathbf{j} \rangle\rangle = -\frac{e\alpha N_e}{2\pi^2 \hbar \omega} \sum_{\mathbf{n}} H^{(0)}_{0,\mathbf{n}} \mathbf{n} \int d\mathbf{k}\, f(\varepsilon(\mathbf{k})) \cos(\mathbf{k}\cdot\mathbf{n}) \times I_{kin}(\varepsilon_1, \varepsilon_2; \varphi), \tag{16}$$

where the dimensionless I_{kin} is

$$I_{kin}(\varepsilon_1, \varepsilon_2; \varphi) = \int_0^{2\pi} d\tau \int_0^{2\pi} d\tau' (\tau - \tau') \times \sin\left[\frac{e}{\hbar c}\mathbf{A}\left(\frac{\tau}{\omega}\right)\cdot\mathbf{n} - \frac{e}{\hbar c}\mathbf{A}\left(\frac{\tau'}{\omega}\right)\cdot\mathbf{n}\right] - 2\pi \int_0^{2\pi} d\tau \int_0^{\tau} d\tau' \times \sin\left[\frac{e}{\hbar c}\mathbf{A}\left(\frac{\tau}{\omega}\right)\mathbf{n} - \frac{e}{\hbar c}\mathbf{A}\left(\frac{\tau'}{\omega}\right)\cdot\mathbf{n}\right]. \tag{17}$$

For simplicity we again adopt the tight-binding approximation below.

Let us study the ε and φ dependencies of the dc current. In the kinetic case there are also symmetries with respect to φ, which can be verified readily from Eqs. (17) and (4). This time, however, the second equation is antisymmetric in contrast to Eq. (10), where antisymmetric was the first one:

$$\langle\langle \mathbf{j}(t) \rangle\rangle|_{-\varphi} = \langle\langle \mathbf{j}(t) \rangle\rangle|_{\varphi}, \quad \langle\langle \mathbf{j}(t) \rangle\rangle|_{\pi-\varphi} = -\langle\langle \mathbf{j}(t) \rangle\rangle|_{\varphi}. \tag{18}$$

The symmetry properties (18) reduce the study to the range $0 < \varphi < \pi/2$. For $\varphi = \pm\pi/2$ the current is identically zero. Note the difference with the dynamic regime, when the current was zero at $\varphi = 0$ instead. Increasing the phase shift φ from 0 to π one obtains the change of the dc electric current $\langle\langle j \rangle\rangle$ from some positive (negative) value to some negative (positive) one, depending on the values of ε_1 and ε_2, see Fig. 2(a). The possibility to control both the directionality and the magnitude of the current in the kinetic regime is a fact, qualitatively similar to the dynamic case. However, this time φ varies in the range $0 < \varphi < \pi$, and quantitatively the corresponding dependencies are drastically different, cf. Figs. 1 and 2.

The $\varepsilon_1, \varepsilon_2$ dependencies of the current $\langle\langle j \rangle\rangle$ in the kinetic regime are depicted in Fig. 2(b) for $\varphi = 0$. Changing either one or both of the parameters $\varepsilon_1, \varepsilon_2$ along a suitable trajectory one can go from positive values of the current through zero to negative values, or vice versa. For example, from the maximal positive value $-I_{kin} = 18.37\ldots$ at $\varepsilon_1 = 2.188\ldots$, $\varepsilon_2 = 1.051\ldots$, $\varphi = 0$ to the minimal negative one $-I_{kin} = -6.730\ldots$ at $\varepsilon_1 = 2.377\ldots$, $\varepsilon_2 = 2.991\ldots$, $\varphi = 0$. Obviously, the decrease of one (or both) component(s) of the applied field may lead to counterintuitive increase of the resulting dc current, Fig. 2(b), and vise versa. In comparison to the dynamic regime the shape of the kinetic surface representing the dc current as function of field amplitudes, however, is drastically different (note different ranges for φ).

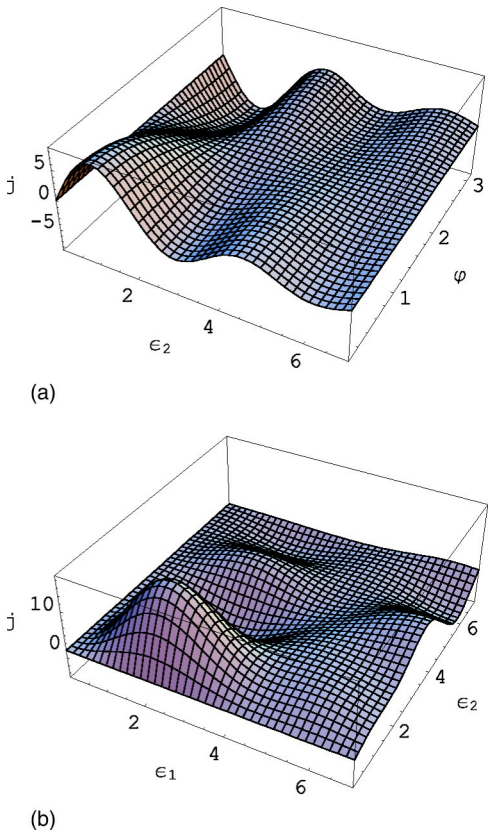

FIG. 2. The dc electric current $\langle\langle j \rangle\rangle$ (arbitrary units) in the kinetic regime (a) as function of the phase shift φ in the range $[0, \pi]$ and of the dimensionless field amplitude ε_2 with $\varepsilon_1 = 1$ fixed, and (b) as function of $\varepsilon_1, \varepsilon_2$ for phase shift $\varphi = 0$.

It can be demonstrated from Eq. (17) that the dc current is zero if either one of the fields ε_1 or ε_2 vanishes. In this respect the situation differs from the dynamic regime when the current was nonzero if $\varepsilon_1 = 0, \varepsilon_2 \neq 0$. The reason is in the fact that at long time relaxation smears out the effect of initial-field value, discussed in the preceding section. Equivalently, it restores the invariance of average quantities with respect to the shift of the time origin. Then in a single applied field nothing breaks the left-right symmetry on average, so that there cannot be any dc current. Obviously, the introduction of the second coherent field breaks that symmetry and the dc current appears—away from the lines $\varepsilon_1 = 0$ and $\varepsilon_2 = 0$.

An explicit expansion can be obtained for $\varepsilon_1 \ll 1, \varepsilon_2 \ll 1$:

$$I_{kin}(\varepsilon_1, \varepsilon_2; \varphi) = -\tfrac{3}{2}\pi^2 \cos\varphi \varepsilon_1^2 \varepsilon_2 + \cdots. \tag{19}$$

It is linear in ε_2, quadratic in ε_1, and becomes zero for $\varphi = \pm\pi/2$, in agreement with Fig. 2(b). The functional dependence of the current (19) on $\varepsilon_1, \varepsilon_2$, and φ agrees with the result of Ref. 23, obtained for a different model of quantum dissipation in the incoherent sequential tunneling regime. The leading term of the dc current, Eq. (19), comes from the third-order nonlinear response[20,21,23] and thus is proportional to the time average of $\mathbf{E}^3(t)$, Eq. (1), with coefficient $\sigma^{(3)}(\omega, \omega, -2\omega)$. Such dependence on electric-field ampli-

tudes has been observed in experiments on coherent control in optical superlattices,[12] semiconductor superlattices,[19] and bulk semiconductors.[21]

IV. DISCUSSION

To summarize, we have considered the coherently controllable dc current (zero-harmonic generation) in an independent-electron one-band conductor in two regimes: dynamic without relaxation (or short time) and kinetic with the account of scattering (long time). The applied field has two components with frequencies ω and 2ω with phase shift φ. Explicit expressions of the dc current are obtained in both cases, with special emphasis in the kinetic case on the low-scattering nearly coherent regime. Coherent control is possible in either one. However, the field- and phase-shift dependencies of the dc current in both cases are drastically different. One of the reasons for that is in the fact that in the dynamic regime the effect of initial-field value (or initial phase shift φ) does not decay in time due to absence of relaxation. In the kinetic long-time regime, on the contrary, the electrons thermalize and the initial conditions play no role. Another important difference is in the fact that the dc current in the kinetic long-time regime arises due to the nonlinear (starting with third-order) response. In the dynamic regime the lowest-order contribution is first order in the field ε_2. Additionally, in the kinetic regime the result depends considerably on the space symmetry of the lattice.

In the kinetic regime the dc current in an ac field often vanishes [for example, in the $\cos(\omega t)$ field, considered most often], unless other coherent components of the applied field break the space inversion symmetry. The same is true for the dynamic regime as well. However, in the latter case there is an additional mechanism for nonzero current formation: coherence effects in electron evolution, including the effect of initial conditions, which do not decay due to absence of relaxation. The latter initial-field-value effect is quite transparent: The initial band filling is symmetric, $\rho_{\mathbf{k},\mathbf{k}}(0) = \rho_{-\mathbf{k},-\mathbf{k}}(0)$, but the field may have nonzero initial value $\mathbf{E}(t=0) \neq 0$, which breaks the symmetry in electron evolution (similar to Refs. 32 and 33).

Coherently controllable dynamic dc currents should be observable on shorter time scales than the ones in the kinetic regime; namely, which one takes place is determined by the time of observation: at short time, comparable to interscattering time the first one is realized, while for long time, exceeding the inverse relaxation rate, the second one should be observed.

The mathematical reason for the cited differences between the dynamic and the low-scattering kinetic regimes is the additional integral over time in Eq. (6) as compared to Eq. (3), which occurs due to the implicit averaging over scattering events, entering the kinetic formulas. This additional integration cancels the contribution, linear in the field due to lattice symmetry in Eq. (14) and thus raises the order of the contributing nonlinear processes.

In the final expressions for the current, Eqs. (8) and (16) within the tight-binding approximation the initial band filling and the equilibrium distribution function factor out from the field and phase dependence, which enables the study of E_1, E_2, and φ dependence of the current in general form without specifying $\rho_{\mathbf{k},\mathbf{k}}(0)$ and $f(\varepsilon(\mathbf{k}))$.

We assumed the carriers to be already present in the band either intrinsically or due to some unspecified mechanism, and focused on the coherent control effect upon intraband evolution. Intraband propagation by itself provides the means for coherent control. However, carrier generation, for example, by photoexcitation from bound states on donors,[17] or via interband transitions,[19,20] can also be incorporated by substituting suitable expressions in place of N_e and $\rho_{\mathbf{k},\mathbf{k}}(0)$, the latter possibly becoming nonsymmetric in $\pm\mathbf{k}$ to incorporate the coherent control effect upon the photoemission process.

The case of injected k electrons in the coherent dynamic regime might be of some interest as well. All the field and phase dependence of the dynamic current in Eq. (8) is incorporated in the factor $I_{dyn}(\varepsilon_1,\varepsilon_2;\varphi)$. Thus, all the injected electrons provide similar dependencies on $\varepsilon_1,\varepsilon_2,\varphi$, which differ, however, by a numerical prefactor, provided by the initial distribution: the sign of the dynamic current will be opposite for electrons injected in the upper and lower parts of the band through $\cos(k)$. This gives the theoretical possibility to find out where spectrally the electrons in the band are injected through the measurement of the sign of the current. If the whole band is populated uniformly, the dynamic current becomes zero. In the thermalized long-time kinetic regime (16) there is no such dependence on initial conditions.

The change of coherent control, i.e., current magnitude and direction, in going from the dynamic to the kinetic regime could serve as a monitor for establishing the presence of these regimes of nonlinear response.

Finally let us address the question of observability of the considered effects in experiments on GaAs/GaAlAs semiconductor superlattices. The relaxation time α^{-1} we assume ~ 1 ps (Ref. 27 at $T=10$ K, Refs. 22 and 28). To meet the slow-scattering condition $\alpha T \ll 1$, the laser frequency should be high enough, $\nu=\omega/2\pi=1/T \gg \alpha=1$ THz, in our case. On the other hand, for the one-band approximation to be valid, the laser photon energy should be smaller than the band gap Δ, $\nu \ll \Delta/h$. For the band gap to be bigger, the superlattice GaAs wells should be narrow enough and the spacer GaAlAs layers should be thick enough (but not too thick in order the overlap does not become too small). For example, in Ref. 19 they are 5.5 nm and 32.5 nm, respectively, so that the band gap is 152 meV. For the interband transitions to be negligible, we assume the upper limiting frequency ~ 10 THz, corresponding to photon energy ~ 40 meV. Thus there should be a narrow range for the laser frequency in between 10^{12} and 10^{13} Hz, where our results for the coherent control through intraband evolution should be valid. The manufacture of bigger band gap superlattices with longer relaxation time (lower-temperature measurements) should increase the range of its applicability.

Let us estimate the peak value of the current in the kinetic regime. For the carrier density $N_e \sim 10^{15}$ cm^{-3}, Refs. 8 and 27, miniband width ~ 2 meV, laser frequency $\nu \sim 5$ THz, field amplitudes $E_1 \sim E_2 \sim 10$ kV/cm, phase shift $\varphi=0$, and

the current density is $\sim 0.3\ \mu A/\mu m^2$, or for a spot size $\sim 50\ \mu m$, Ref. 19, the estimated current is ~ 0.6 mA, well within the range of measurements. In the dynamic regime the analogous current for $\varphi = 0$ is identically zero, as noted above. For $\varphi = \pi/2$ the peak value of the short-time dynamic current density is estimated to be $\sim 1\ \mu A/\mu m^2$.

In optical superlattices the relaxation processes are much slower, so that the conditions for the observation of the considered effects are more favorable. For example, the apparent deviation from the constant velocity at short time in the inset of Fig. 2 of Ref. 12 obviously is due to the stated difference in short-time and long-time response.

The study of dynamic (short-time) regime in semiconductor superlattices requires subpicosecond measurements. For all-optical response the 1 ps–100 fs range is accessible, though such measurements of transient currents pose more problems.[1,2,6–9] In contrast to that, optical superlattices require the time scale of only ~ 1–$10\ \mu$s Ref. 12, which should be quite accessible. In any case, we hope the provided theoretical consideration of the short-time dynamic regime will serve for the clarification of the nontrivial time evolution of the response and coherent control in superlattices.

We believe that the coherent control, exercised through intraband evolution, can be observed at low temperatures in the electromagnetic response of high-quality semiconductor superlattices,[1–9,27,28] optical lattices,[9–12] and quantum wires[6,9] in the mid-THz range.

ACKNOWLEDGMENTS

One of the authors (K.P.) acknowledges financial support from NSERC (Canada), and RFBR (Russia). K.P. also gratefully acknowledges numerous helpful discussions with Professor A. A. Ovchinnikov.

[1] *Coherent Control in Atoms, Molecules, and Semiconductors*, edited by W. Poetz and A. Schroeder (Kluwer Academic, Dordrecht, 1999).
[2] F.G. Bass, and A. Bulgakov, *Kinetic and Electrodynamic Phenomena in Classical and Quantum Semiconductor Superlattices* (Nova Science, Commack, NY, 1997).
[3] R.D. Astumian, Science **276**, 917 (1997).
[4] A.D. Yoffe, Adv. Phys. **50**, 1 (2001).
[5] P. Reimann, Phys. Rep. **361**, 57 (2002).
[6] F. Rossi and T. Kuhn, Rev. Mod. Phys. **74**, 895 (2002).
[7] V.M. Axt and S. Mukamel, Rev. Mod. Phys. **70**, 145 (1998).
[8] A. Wacker, Phys. Rep. **357**, 1 (2002).
[9] M. Gluck, A.R. Kolovsky, and H.J. Korsch, Phys. Rep. **366**, 103 (2002).
[10] M.G. Raizen, C. Salomon, and Q. Niu, Phys. Today **50** (7), 30 (1997).
[11] G. Grynberg and C. Robilliard, Phys. Rep. **355**, 335 (2001).
[12] M. Schiavoni, L. Sanchez-Palencia, F. Renzoni, and G. Grynberg, Phys. Rev. Lett. **90**, 094101 (2003).
[13] A. Tikhonov, R.D. Coalson, and Yu. Dahnovsky, J. Chem. Phys. **116**, 10 909 (2002).
[14] J. Lehmann, S. Kohler, P. Hanggi, and A. Nitzan, Phys. Rev. Lett. **88**, 228305 (2002).
[15] A. Keller, O. Atabek, V. Mujica, and M. Ratner, J. Phys. B **35**, 4981 (2002).
[16] A. D. Bandrauk and H. Kono, in *Advances in Multiphoton Processes and Spectroscopy*, edited by S. H. Lin (World Scientific, Singapore, 2003) Vol. 15.
[17] G. Kurizki, M. Shapiro, and P. Brumer, Phys. Rev. B **39**, 3435 (1989).
[18] P. Brumer and M. Shapiro, Chem. Phys. Lett. **126**, 541 (1986).
[19] E. Dupont, P.B. Corkum, H.C. Liu, M. Buchanan, and Z.R. Wasilewski, Phys. Rev. Lett. **74**, 3596 (1995).
[20] R. Atanasov, A. Hache, J.L.P. Hughes, H.M. van Driel, and J.E. Sipe, Phys. Rev. Lett. **76**, 1703 (1996).
[21] A. Hache, Y. Kostoulas, R. Atanasov, J.L.P. Hughes, J.E. Sipe, and H.M. van Driel, Phys. Rev. Lett. **78**, 306 (1997).
[22] K.N. Alekseev, E.H. Cannon, J.C. McKinney, F.V. Kusmartsev, and D.K. Campbell, Phys. Rev. Lett. **80**, 2669 (1998).
[23] I. Goychuk and P. Hanggi, Europhys. Lett. **43**, 503 (1998).
[24] J. Karczmarek, M. Stott, and M. Ivanov, Phys. Rev. A **60**, R4225 (1999).
[25] S. Flach, O. Yevtushenko, and Y. Zolotaryuk, Phys. Rev. Lett. **84**, 2358 (2000); O. Yevtushenko, S. Flach, and K. Richter, Phys. Rev. E **61**, 7215 (2000); S. Denisov, S. Flach, A.A. Ovchinnikov, O. Yevtushenko, Y. Zolotaryuk, *ibid.* **66**, 041104 (2002).
[26] S. Hughes and D.S. Citrin, Phys. Rev. B **59**, R5288 (1999).
[27] K. Unterrainer, B.J. Keay, M.C. Wanke, S.J. Allen, D. Leonard, G. Medeiros-Ribeiro, U. Bhattacharya, and M.J.W. Rodwell, Phys. Rev. Lett. **76**, 2973 (1996).
[28] T. Meier, G. von Plessen, P. Thomas, and S.W. Koch, Phys. Rev. B **51**, 14 490 (1995).
[29] D.H. Dunlap and V.M. Kenkre, Phys. Rev. B **34**, 3625 (1986); Phys. Lett. A **127**, 438 (1988); Phys. Rev. B **37**, 6622 (1988).
[30] K.A. Pronin, A.D. Bandrauk, and A.A. Ovchinnikov, J. Phys.: Condens. Matter **6**, 4721 (1994); Phys. Rev. B **50**, 3473 (1994); Synth. Met. **69**, 707 (1995); **71**, 1687 (1995); **85**, 1169 (1997); A.A. Ovchinnikov and K.A. Pronin, Chem. Phys. **235**, 93 (1998).
[31] K.A. Pronin, P. Reineker, and A.D. Bandrauk (unpublished).
[32] A.D. Bandrauk and S. Chelkowski, Phys. Rev. Lett. **84**, 3562 (2000).
[33] S. Chelkowski, M. Zamojski, and A.D. Bandrauk, Phys. Rev. A **63**, 023409 (2001).
[34] P. Reineker in *Springer Tracts in Modern Physics* edited by G. Höhler (Springer, Berlin, 1982), Vol. 94, p. 101.