Method for observing Bloch oscillations in the time domain

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We suggest an experimental method which should, at least in principle, be capable of measuring Bloch oscillations directly in the time domain. The method consists of measuring the spontaneous photon-echo signal in a time-resolved four-wave-mixing experiment on a semiconductor superlattice. We calculate the third-order nonlinear polarization of an idealized superlattice in the presence of a homogeneous electric field applied along the growth direction. In the limits of both vanishing and infinite fields, the echo signal produced by the nonlinear polarization should be independent of the delay time τ between the exciting pulses, if irreversible dephasing processes are disregarded. For finite fields, the echo signal should exhibit a modulation periodic in τ, with the periodicity of the Bloch oscillations. If observed experimentally, this would represent a direct manifestation of Bloch oscillations in the time domain.

I. INTRODUCTION

The dynamics of Bloch electrons in the presence of a homogeneous electric field is one of the fundamental problems of the quantum theory of solids. In 1928, Bloch[1] stated that, in the presence of an applied homogeneous field $\mathbf{F}$ a wave packet composed of a superposition of Bloch states and peaked about some quasimomentum $\hbar\mathbf{k}$ moves in $\mathbf{k}$ space, changing its quasimomentum at a time rate proportional to $\mathbf{F}$. Moreover, it can be shown that each individual Bloch state changes its wave vector according to the so-called acceleration theorem[2-5]

$$\dot{\mathbf{k}}=e\mathbf{F}/\hbar \; . \tag{1}$$

Thus, in the absence of interband tunneling and scattering processes, a Bloch electron subjected to a homogeneous electric field moves at a constant velocity in $\mathbf{k}$ space and executes a periodic motion in the reduced-zone scheme (a so-called "Bloch oscillation") by undergoing a Bragg reflection between each two traversals of the Brillouin zone. There are two mechanisms impeding a fully periodic motion: interband tunneling and scattering processes. Interband tunneling is an intricate problem and still at the center of a continuing debate (see Ref. 6, and references therein). It is only during the last decade that upper boundaries for the interband tunneling probability have been established at a rigorous level,[7] which show that an electron may execute a number of Bloch oscillations before tunneling out of the band. The second mechanism impeding a fully periodic motion is scattering off phonons, impurities, etc. This results in lifetimes shorter than the period of the oscillatory motion T_{Bloch} for all reasonable values of the electric field, so that Bloch oscillations should not be observable in conventional solids. In superlattices, however, the situation is more favorable because of the smaller T_{Bloch} times resulting from the small width of the mini-Brillouin zone in one direction. In fact, the stationary-state counterparts of the Bloch oscillations, the Wannier-Stark states, have been observed in semiconductor superlattices.[8,9]

A direct time-resolved experimental detection of Bloch oscillations, however, is still lacking. In this paper, we would, therefore, like to suggest an experimental method which should, at least in principle, be capable of measuring Bloch oscillations directly in the time domain. It consists of measuring the spontaneous photon-echo signal in a time-resolved degenerate four-wave-mixing (DFWM) experiment on a semiconductor superlattice. In a theoretical investigation of photon echo in bulk semiconductors, Zakharov and Manykin[10] found that, for high enough fields, the echo signal should exhibit a characteristic temporal modulation with the periodicity of the Bloch oscillations. Our suggestion is that such a signal modulation should be observable in semiconductor superlattices already at moderate fields, and could thus be used to detect Bloch oscillations in the time domain.

This paper is organized as follows: In Sec. II we will give a description of Bloch oscillations in a one-dimensional lattice. Section III contains a brief summary of some general features of DFWM in the spontaneous photon-echo configuration. In Sec. IV we calculate the photon-echo amplitude of an idealized superlattice in the presence of an electric field applied along the growth direction. In Sec. V we will illustrate our results by means of a simple tight-binding model. Finally, Sec. VI contains a brief discussion of the assumptions made in our analysis, followed by the conclusions.

II. BLOCH OSCILLATIONS IN A ONE-DIMENSIONAL LATTICE

In the following analysis, we will restrict ourselves to the study of a one-dimensional infinite lattice with nondegenerate bands. The lattice is assumed to be regular, i.e., there is no static disorder. Our treatment is based on the one-particle approximation, so both Coulombic effects and quasiparticle interactions are neglected.

In the zero-field case, the eigenvalues of the electronic Hamiltonian are given by the familiar band structure, $\varepsilon_n(k)$, with band index n and wave number k. The eigenstates are the usual Bloch states, $|n,k\rangle$, with spatial representation

$$\langle x|n,k\rangle = e^{ikx}u_{n,k}(x)\ ,\quad u_{n,k}(x+d)=u_{n,k}(x)\ , \tag{2}$$

where d is the lattice period. The time evolution of an electron prepared at time $t=0$ in a state from the nth band can then be calculated using the time-evolution operator

$$\hat{U}_n(t)=\int_{-\pi/d}^{+\pi/d}dk\,\exp\left[-\frac{i}{\hbar}\varepsilon_n(k)t\right]|n,k\rangle\langle n,k|\ . \tag{3}$$

In the case of nonzero field $F\neq 0$ the situation is much more involved due to the perturbation caused by the applied field. This perturbation leads to coupling between k subspaces corresponding to different bands of the zero-field Hamiltonian. However, this difficulty is removed in the treatment given in Ref. 11, in which the zero-field band subspaces are slightly "deformed" to field-dependent band subspaces in such a way that coupling between different bands is minimized. Within each of these field-dependent band subspaces, there exist quasistationary states, which are exponentially localized. We will call these states "Wannier-Stark states" and denote them by $|n,\nu\rangle^F$, where $\nu=0,\pm1,\pm2,\ldots$. The lifetime of these quasibound states is limited by interband tunneling due to the field-induced coupling between different bands. The Wannier-Stark states of each band subspace can be expressed as linear superpositions of Bloch states from the same band subspace $|n,k\rangle^F$:[6]

$$|n,\nu\rangle^F=\left[\frac{d}{2\pi}\right]^{1/2}\int_{-\pi/d}^{+\pi/d}dk\,\exp(i\nu kd)\exp\left[-\frac{i}{eF}\int_{-\pi/d}^{k}[\varepsilon_n^F(k')-\bar{\varepsilon}_n^F]dk'\right]|n,k\rangle^F\ ,\quad \nu=0,\pm1,\pm2,\ldots\ , \tag{4}$$

where $\varepsilon_n^F(k)$ is a real function of k with periodicity $2\pi/d$, tending smoothly to $\varepsilon_n(k)$ as $F\to 0$, and

$$\bar{\varepsilon}_n^F\equiv\frac{d}{2\pi}\int_{-\pi/d}^{+\pi/d}\varepsilon_n^F(k)dk\ . \tag{5}$$

The energies of the Wannier-Stark states of each band subspace form a discrete, ladderlike spectrum, the well-known Wannier-Stark ladder

$$E_{n,\nu}^F=\bar{\varepsilon}_n^F+\nu eFd\ . \tag{6}$$

The first step in our treatment of the Bloch oscillations is to write down an approximative time-evolution operator for the nth band, using the above Wannier-Stark states:

$$\hat{U}_n^F(t)=\sum_\nu\exp\left[-\frac{i}{\hbar}E_{n,\nu}^F t\right]|n,\nu\rangle^{F\,F}\langle n,\nu|\ . \tag{7}$$

$\hat{U}_n^F(t)$ is a good approximation for the time evolution of states of the nth band in a time interval which is determined by the lifetime of the Wannier-Stark states. Since finite lifetime effects due to field-induced interband tunneling will be of importance only for very strong fields, we will disregard them in the following.

We will now write the time-evolution operator of Eq. (7) in a more instructive way. Inserting Eqs. (4)–(6) into Eq. (7) we obtain, after a short calculation,

$$\hat{U}_n^F(t)=\int_{-\pi/d}^{+\pi/d}dk\,\exp\left[-\frac{i}{\hbar}\int_0^t\varepsilon_n^F\left[k+\frac{eF}{\hbar}t'\right]dt'\right]\times\left|n,k+\frac{eF}{\hbar}t\right\rangle^{F\,F}\langle n,k|\ . \tag{8}$$

Note that $\hat{U}_n^F(t)$ tends to $\hat{U}_n(t)$ smoothly as $F\to 0$.

We will now use $\hat{U}_n^F(t)$ to calculate the time evolution of an electron in the nth band. Suppose that at $t=0$, the electron is in a state $|\psi_0\rangle$ composed of a superposition of Bloch states of the nth band. Its state will evolve in time according to

$$|\psi(t)\rangle=\hat{U}_n^F(t)|\psi_0\rangle=\int_{-\pi/d}^{+\pi/d}dk\,\exp\left[-\frac{i}{\hbar}\int_0^t\varepsilon_n^F\left[k+\frac{eF}{\hbar}t'\right]dt'\right]\times\left|n,k+\frac{eF}{\hbar}t\right\rangle^{F\,F}\langle n,k|\psi_0\rangle\ . \tag{9}$$

We see that the wave packet representing $|\psi(t)\rangle$ moves in **k** space within the nth band, each k component acquiring a dynamical phase

$$\exp\left[-\frac{i}{\hbar}\int_0^t\varepsilon_n^F\left[k+\frac{eF}{\hbar}t'\right]dt'\right]\ .$$

After a time $T_{\text{Bloch}}=2\pi\hbar/eFd$, the electron reaches an equivalent point in **k** space and, due to the conservation of the band index n and the periodicity of $\varepsilon_n^F(k)$, the initial state $|\psi_0\rangle$ is restored except for a possible overall phase factor, which is of no importance. In the reduced-zone scheme, the electron has thus executed a Bloch oscillation.

In the particular case of an electron prepared in a Bloch state, say $|n,k_o\rangle^F$, Eq. (9) yields immediately

$$\hat{U}_n^F(t)|n,k_0\rangle^F=\exp\left[-\frac{i}{\hbar}\int_0^t \varepsilon_n^F\left[k_0+\frac{eF}{\hbar}t'\right]dt'\right]\times\left|n,k_0+\frac{eF}{\hbar}t\right\rangle^F , \quad (10)$$

which repeats the statement made in the acceleration theorem, Eq. (1).

Alternatively, we may use $\hat{U}_n^F(t)$ to calculate the time evolution of an operator in the Heisenberg picture. In our further considerations, we will be particularly interested in the time evolution of the electric dipole-moment operator, $\hat{P}(t)$. We therefore calculate the matrix element of $\hat{P}(t)$ between Bloch states of different bands

$$^F\langle n',k'|\hat{P}(t)|n,k\rangle^F={}^F\langle n',k'|\hat{U}_n^{F\dagger}(t)\hat{P}\hat{U}_n^F(t)|n,k\rangle^F =\delta(k'-k)P_{nn'}^F(k,t) , \quad (11)$$

where

$$P_{nn'}^F(k,t)\equiv p_{nn'}^F\left[k+\frac{eF}{\hbar}t\right]\exp[-i\phi_{nn'}^F(k,t)] . \quad (12)$$

$p_{nn'}^F(k)$ is the usual interband optical matrix element; the phase angle $\phi_{nn'}^F(k,t)$ is given by

$$\phi_{nn'}^F(k,t)\equiv\frac{1}{\hbar}\int_0^t\left[\varepsilon_n^F\left[k+\frac{eF}{\hbar}t'\right]-\varepsilon_{n'}^F\left[k+\frac{eF}{\hbar}t'\right]\right]dt' . \quad (13)$$

In Eq. (11), we have made the standard approximation in which the photon momentum is neglected so that $P(t)$ couples Bloch states of the same k.

Looking at Eqs. (11)–(13), we see that the time evolution of the dipole matrix elements mirrors the dynamics of the Bloch electron described in Eq. (9), including the occurrence of Bloch oscillations. The idea of the experiment suggested in this paper is to detect Bloch oscillations by measuring the dynamical phases $\exp[-i\phi_{nn'}^F(k,t)]$.

III. SOME FEATURES OF SPONTANEOUS PHOTON ECHO

This part of the paper summarizes some features of time-resolved DFWM in the spontaneous photon-echo configuration. In such an experiment a coherent optical polarization is produced in the sample, say, at a time $t=0$, by a short laser pulse with wave vector $\mathbf{k}_1$. This coherent polarization develops in time. After a time delay τ a second pulse with wave vector $\mathbf{k}_2$ is applied. In the case of an inhomogeneously broadened transition, a spontaneous photon-echo signal is then emitted in the direction $\mathbf{k}_3\equiv2\mathbf{k}_2-\mathbf{k}_1$ at a time 2τ. The signal is monitored as a function of the delay time τ.

The following paragraphs provide some equations which will be needed in the subsequent sections of this paper. The component of the optical polarization giving rise to the emitted photon-echo signal may be written as

$$P_{\mathbf{k}_3}(\mathbf{r},t,\tau)=\tilde{P}(\mathbf{k}_3,t,\tau)e^{i(\mathbf{k}_3\cdot\mathbf{r}-\omega_L t)}+\text{c.c.} , \quad (14)$$

where ω_L is the central frequency of the laser pulses. Assuming optically thin samples, the intensity of the emitted signal is given by

$$I(t,\tau)=|\tilde{P}(\mathbf{k}_3,t,\tau)|^2 . \quad (15)$$

The computation of $\tilde{P}(\mathbf{k}_3,t,\tau)$ is achieved most easily for the case of very short laser pulses, idealized as δ pulses, i.e., having amplitudes

$$\tilde{E}_1(t)=A_1\delta(t) \text{ and } \tilde{E}_2(t)=A_2\delta(t-\tau) . \quad (16)$$

Making use of the nonlinear response function of Ref. 12, it can be shown, for a medium characterized by a time-independent Hamiltonian, that $\tilde{P}(\mathbf{k}_3,t,\tau)$ is approximately given by

$$2i\,A_1^*A_2^2e^{i\omega_L(t-\tau)}\langle\text{Tr}[\hat{P}\hat{U}(\tau-t)\hat{P}\hat{U}(t-\tau)\hat{P}\hat{U}(\tau)\hat{\rho}(-\infty)\hat{P}\hat{U}(-\tau)]\rangle_{\text{conf}}\equiv\tilde{P}_e(\mathbf{k}_3,t,\tau) , \quad (17)$$

where $\hat{\rho}(t)$ is the density operator, $\hat{U}(t)$ is the time evolution operator of the medium, and a configurational averaging is included. $\tilde{P}_e(\mathbf{k}_3,t,\tau)$ is an excellent approximation of $\tilde{P}(\mathbf{k}_3,t,\tau)$ except in a time interval around $t=0$ which is roughly given by the inverse width of the inhomogeneous broadening of the optical transition.

If the inhomogeneous broadening is large enough $\tilde{P}_e(\mathbf{k}_3,t,\tau)$ will be sharply peaked at $t=2\tau$, and negligible for all times $t\neq2\tau$. The time-integrated signal then shows the following dependence on τ:

$$\int_0^\infty I(t,\tau)dt\propto|\tilde{P}_e(\mathbf{k}_3,\,t=2\tau,\,\tau)|^2 , \quad (18)$$

except in the aforementioned time interval; in other words, the time-integrated signal is given by relation (18) for all delay times τ greater than the inverse width of the inhomogenous broadening. We will call $\tilde{P}_e(\mathbf{k}_3,\,t=2\tau,\,\tau)$ "echo amplitude."

IV. SPONTANEOUS PHOTON ECHO IN A SUPERLATTICE

In this part of the paper, we will apply the results of the preceding two sections to the study of DFWM in a semiconductor superlattice in the presence of a static electric field applied along the growth direction of the superlattice. We consider an idealized superlattice in which the component of the electronic quasimomentum perpendicular to the applied field, $\hbar\mathbf{k}_\perp$, is independent of the field. The problem is then reducible to that of a one-dimensional lattice, except for a band dispersion in $\mathbf{k}_\perp$ direction, which we may treat as an inhomogeneous broadening of the transition frequency. In the absence of band degeneracies, we are free to use the results of Sec. II for the calculation of the third-order polarization amplitude $\tilde{P}_e(\mathbf{k}_3,t,\tau)$. The time-integrated DFWM signal can then be calculated by inserting $\tilde{P}_e(\mathbf{k}_3,t,\tau)$ into relation (18).

For simplicity, we will restrict ourselves to the case of zero temperature, where all conduction miniband states are empty and all valence miniband states are occupied. The density operator of Eq. (17), $\hat{\rho}(-\infty)$, is then given by the projection operator upon the latter ones. Inserting $\hat{\rho}(-\infty)$ and complete sets of Bloch states into Eq. (17), one obtains for $\tilde{P}_e(\mathbf{k}_3,t,\tau)$

$$\tilde{P}_e(\mathbf{k}_3,t,\tau)=2i\,A_1^*A_2^2e^{i\omega_L(t-\tau)} \times\left\langle \sum_{\substack{v,v',\\c,c'}}\int_{-\pi/d}^{+\pi/d}dk\,P_{vc}^F(k,-\tau)P_{cv'}^F(k,0) \times P_{v'c'}^F(k,t-\tau) \times P_{c'v}^F(k,0)\right\rangle_{\text{conf}}, \qquad (19)$$

where the sum runs over all valence and conduction miniband (summation indices v, v', c, and c', respectively) and the configurational averaging takes into account the inhomogeneous broadening due the band dispersion in $\mathbf{k}_\perp$ direction; the dipole matrix elements P_{vc}^F are of the kind already encountered in Eq. (12).

Before analyzing the case $F\neq0$, it will be instructive to reconsider some well-known results for the field-free case. For $F=0$, the time evolution between the first and second laser pulse is described by the dipole matrix element $P_{vc}^F(k,-\tau)$ in Eq. (19), with $F=0$. The phase angle of the matrix element is given by

$$\phi_{vc}^{F=0}(k,-\tau)=\frac{1}{\hbar}\int_0^{-\tau}[\varepsilon_c(k)-\varepsilon_v(k)]dt'\ . \qquad (20)$$

Thus, for each k, the corresponding dipole matrix element evolves between times $t=0$ and τ with an effective phase velocity given by $-(1/\hbar)[\varepsilon_c(k)-\varepsilon_v(k)]$, resulting in a reversible dephasing of the matrix elements associated with different values of k. The time evolution of the phase after the second pulse is described by

$$\phi_{vc}^{F=0}(k,t-\tau)=\frac{1}{\hbar}\int_0^{t-\tau}[\varepsilon_c(k)-\varepsilon_v(k)]dt'\ , \qquad (21)$$

i.e., the corresponding dipole matrix element evolves with a phase velocity given by $+(1/\hbar)[\varepsilon_c(k)-\varepsilon_v(k)]$. This means that the nonlinear interaction with the second pulse effectively reverses the direction of the phase motion in the complex plane. Now, for $v=v'$ and $c=c'$ in Eq. (19), the initial phase will be recovered at $t=2\tau$ so that at around $t=2\tau$, the dipole matrix elements of all k move again in phase, giving rise to an echo signal; the echo amplitude is independent of τ, as can be seen by inserting Eqs. (20) and (21) into Eq. (19). For $v\neq v'$ and/or $c\neq c'$, in contrast, the magnitudes of the phase velocities before and after the second pulse will, in general, be different so that no complete rephasing occurs; the corresponding terms in Eq. (19) will, therefore, not contribute to the echo amplitude and shall be neglected in the following. Note that, under this condition, the band structure of the superlattice can be imagined as an ensemble of noninteracting two-level systems in $\mathbf{k}$ space, whose transition frequencies are inhomogeneously broadened due to the miniband dispersion. The τ independence of the echo amplitude is then easily understood as that characteristic of any ensemble of noninteracting inhomogeneously broadened two-level systems.

For $F\neq0$, the situation is more involved than in the field-free case, due to the fact that the integrand in the phase of the dipole matrix elements is now time dependent. Some insight into the time evolution is obtained from a Taylor expansion of the phase angles:

$$\phi_{vc}^F(k,-\tau)=-\frac{1}{\hbar}[\varepsilon_c^F(k)-\varepsilon_v^F(k)]\tau +\frac{1}{eF}O\left[\left[-\frac{eF}{\hbar}\tau\right]^2\right], \qquad (22a)$$

$$\phi_{v'c'}^F(k,t-\tau)=+\frac{1}{\hbar}[\varepsilon_c^F(k)-\varepsilon_v^F(k)](t-\tau) +\frac{1}{eF}O\left[\left[\frac{eF}{\hbar}(t-\tau)\right]^2\right]. \qquad (22b)$$

It is obvious that for small enough τ, i.e., for $\tau\ll T_{\text{Bloch}}$, the phase evolution in the time intervals, $0\le t\le\tau$ and $\tau\le t\le 2\tau$ is characterized by effective phase velocities

$$-(1/\hbar)[\varepsilon_c^F(k)-\varepsilon_v^F(k)]$$

and

$$+(1/\hbar)[\varepsilon_c^F(k)-\varepsilon_v^F(k)]\ .$$

Therefore, on this time scale, the evolution of the polarization is, to a good approximation, analogous to that of the field-free case so that the echo amplitude is at a maximum for small τ. However, with increasing time delay τ, the nonlinear terms in the expansions of the phase angles cease to be negligible, resulting in a time evolution after $t=\tau$ which is not simply the reverse evolution of the time interval between $t=0$ and τ. Consequently, there will be no complete phase recovery at $t=2\tau$. This implies that with increasing τ, the initial maximum of the echo amplitude should be followed by a decay due to the field-induced loss of time reversibility in the phase evolution.

We will now consider the polarization for $F\neq0$ on a larger time scale, i.e., on the time scale of a full Bloch oscillation. From Sec. II, we know that the time evolution of the dipole matrix elements is periodic in time, with period T_{Bloch}. Hence, if the delay time τ is chosen to be equal to T_{Bloch}, all matrix elements will regain their initial phases at $t=\tau$, and again at $t=2\tau$; a maximum in the echo amplitude should then be observed. Increasing τ slightly beyond $T_{\rm Bloch}$ should again decrease the echo amplitude, in complete analogy to the case $\tau \ll T_{\rm Bloch}$ presented above.

Due to the periodicity of the time evolution, the same behavior is expected in the more general case that τ is chosen to be an integral multiple of $T_{\rm Bloch}$, i.e., $\tau = nT_{\rm Bloch}$, $n=1,2,\ldots$. The echo amplitude is thus expected to exhibit a modulation periodic in τ, with peaks at $\tau=0$ and $\tau=nT_{\rm Bloch}$.[13]

Note that, for very strong fields, the prefactor of the nonlinear terms in Eq. (22), $1/eF$, is small; the nonlinear terms will then contribute little to the phase evolution. Therefore, in this case, the echo amplitude will be only weakly modulated as a function of τ; in other words, it will deviate little from its maximum value. This is not surprising, since for very strong fields, the Wannier-Stark states of Eq. (4) are completely localized in each well of the superlattice. The superlattice can then again be regarded as consisting of an ensemble of noninteracting inhomogeneously broadened two-level systems (here, in real space), with the constant echo amplitude which is characteristic of such ensembles. This is in full analogy to the case of photon echo in disordered semiconductors, where strong Anderson localization due to strong disorder gives rise to an echo amplitude almost independent of τ.[14]

In summary, for both $F=0$ and $F\to\infty$, the echo amplitude should be independent of the delay time τ between pulses; for finite fields, it should peak at $\tau = nT_{\rm Bloch}$, $n=0,1,\ldots$. According to Eq. (18), this periodic modulation of the echo amplitude should be experimentally observable as a periodic modulation of the time-integrated DFWM signal.[15] Since the modulation of the expected signal is a consequence of the periodic time evolution of the quantum-mechanical system in the presence of an applied field, it would, if observed, constitute a direct experimental manifestation of Bloch oscillations in the time domain.

V. PHOTON ECHO IN TIGHT-BINDING MINIBANDS

In this part of the paper, we will illustrate the results obtained in the preceding section by specializing them to the model case of simple tight-binding bands. For the idealized superlattice studied in Sec. IV we make the following additional model assumptions.

(1) The band structure of a particular pair of minibands, say, of indices v_0 and c_0, respectively, is of the tight-binding type in the direction parallel to the electric field; more precisely, the reduced (one-dimensional) miniband structure is given by

$$\varepsilon_{v_0}^F(k)=\overline{\varepsilon}_{v_0}^{F=0}+(\Delta_{v_0}/2)\cos(kd)\ , \tag{23a}$$

$$\varepsilon_{c_0}^F(k)=\overline{\varepsilon}_{c_0}^{F=0}-(\Delta_{c_0}/2)\cos(kd)\ , \tag{23b}$$

where $\Delta_{v_0},\Delta_{c_0}>0$ are the zero-field valence and conduction miniband widths, respectively.

(2) $p_{v_0c_0}^F(k)$ is assumed to be independent of k and F.

Note that in assumption (1), we have approximated $\varepsilon_{v_0}^F(k)$ and $\varepsilon_{c_0}^F(k)$ by their zero-field values. This seems to be a good approximation, at least for all qualitative purposes; models with similar miniband structures have been successfully employed in both optics and transport in superlattices.[16,17] By inserting Eq. (23) into Eq. (19), one obtains for the third-order polarization amplitude of the single pair of minibands:

$$P_e(\mathbf{k}_3,t,\tau)=2i\,A_1A_2^2e^{i\omega_L(t-\tau)}p_{v_0c_0}^4J_0\left\{\frac{\Delta_{v_0}+\Delta_{c_0}}{eFd}\left[1+\cos^2\left[\frac{eFd}{2\hbar}t\right]-2\cos\left[\frac{eFd}{2\hbar}t\right]\cos\left[\frac{eFd}{2\hbar}(t-2\tau)\right]\right]^{1/2}\right\}$$
$$\times\left\langle\exp\left[-\frac{i}{\hbar}(\overline{\varepsilon}_{c_0}-\overline{\varepsilon}_{v_0})\right](t-2\tau)\right\rangle_{\rm conf}\ , \tag{24}$$

where J_0 is the Bessel function of index zero. According to relation (18), the time-integrated DFWM signal is given by the square of the modulus of the echo amplitude.[15] In the present case, this is

$$|\widetilde{P}_e(\mathbf{k}_3,\,t=2\tau,\,\tau)|^2=\left|2A_1^*A_2^2p_{v_0c_0}^4J_0\left\{\frac{\Delta_{v_0}+\Delta_{c_0}}{eFd}\left|1-\cos\left[\frac{eFd}{\hbar}\tau\right]\right|\right\}\right|^2 . \tag{25}$$

To evaluate Eq. (25), we take $\Delta_{v_0}+\Delta_{c_0}=70$ meV as a typical value for the sum of the two miniband widths; eFd, i.e., the energy difference between adjacent "steps" of the corresponding Wannier-Stark ladder, is the parameter describing the field strength.

As $F\to 0$,

$$J_0\left\{\frac{\Delta_{v_0}+\Delta_{c_0}}{eFd}\left|1-\cos\left[\frac{eFd}{\hbar}\tau\right]\right|\right\}\simeq J_0\left\{\frac{\Delta_{v_0}+\Delta_{c_0}}{2eFd}\left[\frac{eFd}{\hbar}\tau\right]^2\right\}\to 1\ , \tag{26}$$

i.e., in the limit of vanishing field,

$$|\tilde{P}_e(\mathbf{k}_3, t=2\tau, \tau)|^2$$

is independent of τ [Fig. 1(a)], as stated in Sec. IV. Figure 1(a) also comprises a plot of

$$|\tilde{P}_e(\mathbf{k}_3, t=2\tau, \tau)|^2$$

for $eFd=5$ meV. In this case, the initial maximum in the echo amplitude at $\tau=0$ is followed by a decay due to the field-induced loss of time reversibility in the phase evolution. The field-induced periodic rephasing of the dipole matrix elements can be seen as peaks at $\tau=nT_{\text{Bloch}}$, $n=1,2,\ldots$, each peak corresponding to one Bloch oscillation. Varying the applied field results in changing the time period of the Bloch oscillations; this is illustrated by a comparison of Fig. 1(a) with Fig.1(b), where $eFd=10$ meV. Finally, the weak (albeit fast) modulation of $\tilde{P}(\mathbf{k}_3, t=2\tau, \tau)$ predicted in Sec. IV for very strong fields can be seen in Fig 1(c), where $eFd=160$ meV.

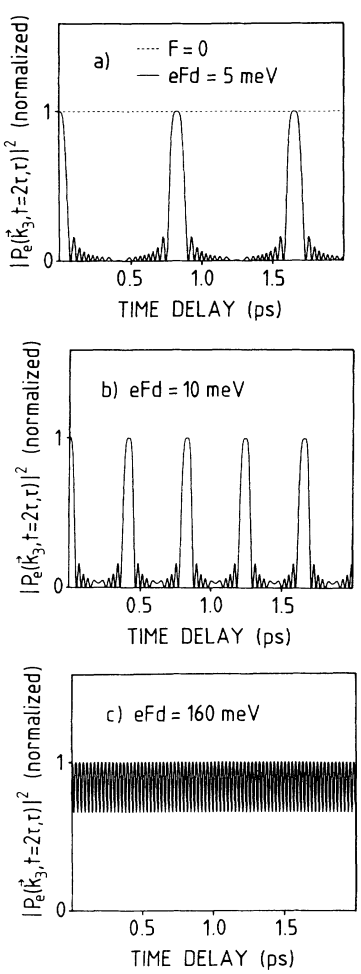

FIG. 1. Square of the modulus of the calculated echo amplitude (normalized to unity) as a function of the delay time τ between pulses, for different strengths of the electric field F applied along the growth direction of the tight-binding superlattice described in Sec. V.

The important point is that Bloch oscillations can be seen clearly in the figures and that they follow the behavior qualitatively described in Sec. IV for the more general case. The analysis within the framework of the simple tight-binding model presented here thus nicely illustrates the results of the preceding section.

VI. DISCUSSION AND SUMMARY

The theoretical treatment presented in this paper has a number of limitations which will be discussed in the following. Some of them are inherent in the one-particle approximation, on which all studies of the Bloch oscillation problem are based; in particular, this approximation disregards Coulombic effects. However "unrealistic" this approach may be, it has led to the (correct) prediction of the existence of Wannier-Stark ladders, in superlattices, and may, therefore, also correctly predict the existence of the time-domain counterpart of the Wannier-Stark ladder, i.e., the Bloch oscillations.

The present analysis of DFWM in a superlattice also neglects irreversible dephasing due to quasiparticle interactions, disorder, and intersubband tunneling. Although the predicted signal modulation may be somewhat masked by these processes, it should still be observable; actually, a similar signal modulation has been observed in the related case of a DFWM experiment on an asymmetric double quantum well.[18]

Another caveat concerns the use of δ pulses in the present analysis. This is a standard approximation in photon-echo calculations, which is useful for avoiding the heavy numerical computation work necessary for consideration of finite pulse widths. Usually, this approximation is justified if the optical pulses are short on the time scale of the dynamical evolution of the system. For the present case, this means that the experimental pulse widths have to be small in comparison with one Bloch period. Additional problems may arise if the optical pulses are spectrally narrow so that they interact resonantly only with part of the states of a given band. We are presently undertaking work to clarify this matter; our first results in this direction indicate that taking into account laser pulses of realistic temporal and spectral widths will not significantly alter the conclusions of the present analysis.[19]

In summary, it has been the aim of this paper to suggest an experimental method which should, at least in principle, be capable of measuring Bloch oscillations directly in the time domain. The experiment suggested here consists of measuring the spontaneous photon-echo signal in a time-resolved DFWM expriment on a semiconductor superlattice. We started with a description of the phenomenon of Bloch oscillations, basing our description on the results obtained by A Nenciu and G. Nenciu for the problem of Bloch electrons in a static electric field. We then applied these results to calculate the third-order nonlinear polarization of an idealized superlattice in the presence of a homogeneous electric field applied along the growth direction. In the limits $F=0$ and $F\rightarrow\infty$, the echo amplitude should be independent of the delay time τ between pulses. For finite fields, the DFWM signal should exhibit echo beats periodic in the time delay τ between the pulses, with the periodicity of the Bloch oscillations. If observed experimentally, they would represent a direct manifestation of Bloch oscillations in the time domain.

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