Electromagnetic emission from a superlattice during ultrafast carrier excitation under the conditions of a Wannier–Stark ladder

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When a superlattice in a uniform electric field is excited by ultrashort pulses, the shape of the emission signal is determined primarily by the relation between the pulse length and the reciprocal step of the Wannier–Stark ladder. A frequency detuning does not play a fundamental role. © *1995 American Institute of Physics.*

Imposing a static, uniform electric field on a semiconductor results in the Franz–Keldysh effect[1,2] in optical absorption (there is a redshift of the absorption edge, and the absorption becomes an oscillatory function of the strength of the static electric field). This effect occurs in the case of steady-state illumination. Effects of a new type arise when carriers are excited by ultrashort femtosecond-range pulses whose length is shorter than the time scales of inelastic processes.[3,4] It has been demonstrated experimentally that excitation results in emission by nonequilibrium carriers in the submillimeter (terahertz) range.[3,4] The temporal shape of the emission signals turns out to be quite different in the cases in which the frequency of the exciting pulse lies above and below the fundamental absorption edge in the semiconductor (in the absence of an electric field).[4] The physical cause of this emission can be summarized as follows. At a frequency larger than the band gap, real carriers are excited; they exist for times on the order of the length of the exciting pulse. A static electric field displaces carriers in different directions, giving rise to a time-dependent dipole moment. This dipole moment in turn gives rise to an emission in the manner of a Hertz vibrator.[5] A time-dependent transport current also arises in this case. If the frequency of the light is instead smaller than the band gap, virtual carriers arise in the band gap in a uniform electric field because of the tails on the densities of states. The virtual carriers cause a displacement current instead of a transport current.[6]

The discussion above pertains to the weak-field limit, where "weak" means that the spectrum of electrons in the bands remains quasicontinuous, and the bands do not "break up" into discrete levels (a Wannier–Stark ladder). Because of the quasicontinuous nature of the spectrum, the dipole moment decays over time after the exciting pulse, even in the absence of inelastic processes. The "strong" fields in which the bulk bands would break up are unattainable in reality. A Wannier–Stark quantization can easily be arranged in superlattices, because of the small width of the miniband. The discrete nature of the spectrum in the case of a Wannier–Stark ladder leads to fundamentally new aspects of the emission. If the pulse length is shorter than the reciprocal of the distance between levels in the ladder, the shape of the response signal depends on only the shape of the exciting pulse, not on the sign of the frequency detuning. If the pulse length is on the order of the reciprocal of the distance between levels, then the emission signal does not decay, in contrast with the behavior in the weak-field case (we are of course assuming that there are no inelastic processes). Along this approach, the limits of weak and strong fields can be considered from a common standpoint.

The tight-binding approximation[7] is convenient for describing the spectrum in a superlattice in a uniform electric field. The reason is that the limits of weak and strong fields can be treated in a unified way. The effective-mass approximation is sufficient in a weak field. In this case the eigenfunctions are Airy functions. [1,2,6] In a strong field, under conditions such that the bands "break up," the effective-mass approximation is not adequate; all states in the original band should be taken into account. This can be done by using the tight-binding method. For our purposes it is sufficient to use the simplest version of the tight-binding method.

We assume that there is one level in each isolated well in the conduction band (c) and the valence band (v). We write the wave function in the form

$$\Psi_{\alpha\nu}(x)=\sum_n A_{\alpha n\nu}\phi_\alpha(x-na), \tag{1}$$

where n is the index of the well; $\alpha=c,v$ is the band index; $\phi_\alpha(x-na)$ is the wave function in an individual well; $A_{\alpha n\nu}$ is the amplitude in well n incorporating overlap with the neighboring wells for eigenstate ν in the superlattice; and a is the period of the superlattice. The Hamiltonian in the tight-binding basis in (1) is

$$H=\sum_{\alpha=c,v}(\varepsilon_{0\alpha}+eaEn)c^+_{\alpha n}c_{\alpha n}+\sum_{\alpha=c,v}t_\alpha(c^+_{\alpha n}c_{\alpha n-1}+c^+_{\alpha n}c_{\alpha n+1}), \tag{2}$$

where the operators $c^+_{\alpha n}$ are creation operators in well n, $t_{c,v}$ is the overlap integral for the overlap between wells, and the term $eaEn$ describes the shift of the levels in the wells in a uniform electric field. In the absence of an electric field the spectrum in the superlattice is

$$\varepsilon_{c,v}=\pm\frac{\varepsilon_g}{2}+t_{c,v}\cos(ka).$$

It follows that t_c and t_v have opposite signs.

The eigenfunctions are found from the recurrence relations

$$A_{c,vn\nu}(\varepsilon_{c,v\nu}-\varepsilon_{0c,v}-eaEn)-t_{c,v}(A_{c,vn+1\nu}+A_{c,vn-1\nu})=0, \tag{3}$$

which are the same as the recurrence relations for Bessel functions,

$$A_{c,vn\nu}=J_{n-\nu}\left(\frac{2t_{c,v}}{eaE}\right). \tag{4}$$

The corresponding eigenvalues of Eq. (3) are

$$\varepsilon_{c,v\nu}=\varepsilon_{0c,v\nu}+\nu eaE. \tag{5}$$

The index ν takes on integer values, as follows from the conditions for the localization of the wave functions at sites in the limit $t \to 0$. In weak fields, $f_{c,v}=2t_{c,v}/eaE \gg 1$, the spectrum is quasicontinuous: $|\varepsilon_\nu - \varepsilon_{\nu\pm 1}|/2t_{c,v} = 1/f_{c,v} \ll 1$. The distance between levels is much smaller than the width of the miniband. The wave functions are delocalized in the miniband. From the asymptotic expressions for the Bessel functions of large argument we find[8]

$$A_{c,vn\nu} \approx \sqrt{\frac{2}{f_{c,v}}} \cos\left(f_{c,v} - (n-\nu)\frac{\pi}{2} - \frac{\pi}{4} \right). \tag{6}$$

In strong fields, $f_{c,v}=2t_{c,v}/eaE \ll 1$, the spectrum consists of discrete levels, separated by distances greater than the width of the original miniband: $|\varepsilon_\nu - \varepsilon_{\nu\pm 1}|/2t_{c,v} = 1/f_{c,v} \gg 1$. The wave functions of the states are localized in the wells from which these states originated:

$$A_{c,vn\nu} \approx \frac{1}{\Gamma(|n-\nu|+1)} \exp[-|n-\nu| \ln(2/|f_{c,v}|)]. \tag{7}$$

The amplitudes fall off more rapidly than exponentially as a function of the site index.

The interaction with an electromagnetic field is described by the Hamiltonian

$$H_{\rm int} = \sum_{\nu,\lambda} E(t)[d_{\nu\lambda} c^+_{c\nu} c_{v\lambda} + \text{H.a.}], \tag{8}$$

where the dipole moment is

$$d_{\nu\lambda} = \int \Psi_{c,nu}(x) \nabla \Psi^*_{vn'\lambda} dx = d_0 \sum_{n,n'} A_{n\nu}(f_c) A_{n'\lambda}(f_v), \tag{9}$$

where

$$d_0 = \delta_{n,n'} \int \phi_c(x-na) \nabla \phi^*_v(x-na) dx.$$

The expression for the dipole moment can be simplified by incorporating the completeness conditions for the Bessel functions,[9]

$$\sum_{n=-\infty}^{+\infty} J_{n-\nu}(\alpha) J_n(\beta) = J_\nu(\alpha-\beta).$$

The collisionless dynamics of the carriers is described by the evolution of the density matrix,

$$i\frac{\partial \hat{\rho}(t)}{\partial t} = [H + H_{\rm int}(t), \hat{\rho}(t)], \tag{10}$$

$$\hat{\rho}(t) = \begin{pmatrix} n_{c,\nu\lambda} & n_{cv,\nu\lambda} \\ n_{vc,\lambda\nu} & n_{v,\nu\lambda} \end{pmatrix}, \quad n_{\alpha\beta,\nu\lambda} = \langle c^+_{\alpha\nu} c_{\beta\lambda} \rangle.$$

We assume that the field has a time dependence

$$E(t) = f(t)\exp(i\omega t) + \text{H.a.}, \tag{11}$$

where $f(t)$ is the shape of the pulse, and ω is the carrier frequency. For conduction electrons a solution can be derived as in Ref. 6; it is

$$n_{c,\nu\lambda}(t)=(E_0d_0)^2\int_{-\infty}^{t}dt'f(t')\exp i[(\varepsilon_{c,\nu}-\varepsilon_{c,\lambda})(t'-t)], \tag{12}$$

$$\sum J_{\lambda-k}(f_{cv})J_{k-\nu}(f_{cv})[F^*(t',\varepsilon_{c,\lambda}-\varepsilon_{v,k}-\Delta)+F(t',\varepsilon_{c,\nu}-\varepsilon_{v,k}-\Delta)],$$

where

$$f_{cv}=f_c-f_v;\ \ \varepsilon_{c,v,k}=eaEk;\ \ \Delta=\omega-\varepsilon_g,$$

$$F(t,\varepsilon)=\exp(i\varepsilon t)\int_{-\infty}^{t}f(\tau)\exp(-i\varepsilon\tau)d\tau=\exp(i\varepsilon t)F'(t,\varepsilon).$$

There are corresponding equations for the holes.

In a weak field, this approach leads to results analogous to those of Ref. 6 $(f_{c,v}\gg 1)$. In addition, there can be cases in which the field is weak for electrons but strong for holes. A situation of this sort is indeed typical of GaAs-based superlattices, because of the difference between the masses of the electrons and holes (in the approach of this paper, this situation corresponds to $f_c\gg 1$ and $f_v\ll 1$ with $m_{c,v}\propto 1/t_{c,v}$) (Ref. 10). An intermediate case of this sort should also be classified as the weak-field case. It can be seen from this approach that the dynamics of the transitions is governed by the lighter particles (the argument of the Bessel functions is determined by the larger of $f_{c,v}$, max $\{f_c,f_v\}$). The physical meaning here is that transitions from a level of the Wannier–Stark ladder in the v band occur to the quasicontinuous spectrum in the c band, and this is the governing effect.

Some new aspects arise when quantization occurs in both bands.

The dipole moment per cell of the superlattice can be written

$$D(t)=ea\sum_{\nu\lambda}[J_\nu(f_c)n_{c,\nu\lambda}J_\lambda(f_c)-J_\nu(f_v)n_{v,\nu,\lambda}J_\lambda(f_v)]. \tag{13}$$

By virtue of translational invariance, the charge in the band is also invariant under a shift:

$$\rho_{\alpha,nn}(t)=e\sum_{\nu\lambda}J_{n-\nu}(f_\alpha)n_{\alpha,\nu\lambda}J_{n-\lambda}(f_\alpha)=e\sum_{\nu\lambda}J_\nu(f_\alpha)n_{\alpha,\nu\lambda}J_\lambda(f_\alpha). \tag{14}$$

A dipole moment arises because of coherent transitions out of the given well in the valence band into the nearest-neighbor wells in the conduction band. Because of the rapid decay of the Bessel functions as a function of the index, it is sufficient to retain simply the terms J_0 and $J_{\pm 1}$ at small values of the argument. It is not sufficient to consider only the terms with $\nu=\lambda=0$. The physical reason is that in (12) these terms describe transitions from the valence band to the conduction band which are vertical transitions in real space; such transitions do not contribute to the dipole moment. The terms $n_{c,v0\pm 1}$ describe transitions from well 0 in the valence band to neighboring wells with indices ± 1 in the conduction band (Fig. 1). This correlated excitation of carriers at different points in coordinate space gives rise to a time-dependent dipole moment. For a given

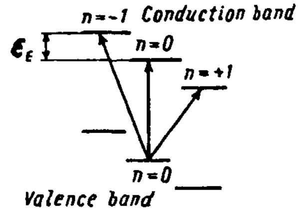

FIG. 1.

frequency, the temporal dynamics of the electrons in the well on the left ($n=-1$) and that on the right ($n=1$) is also different, because of the different frequency detunings. As a result, the total dipole moment does not cancel out. Also using (12), we find

$$D(t)\approx ea(f_{cv}E_0d_0)^2\mathrm{Re}\left\{\int_{-\infty}^{t} f(t')dt'\exp[-i\varepsilon_E(t'-t)]\right.$$
$$\left.\times\,[\mathrm{Re}(F(t',-\Delta)+F(t',\varepsilon_E-\Delta))+F^*(t',-\varepsilon_E-\Delta)+F^*(t',2\varepsilon_E-\Delta)]\right\}, \tag{15}$$

where $\varepsilon_E=eaE$ is the spacing in the Wannier–Stark ladder. If the pulse length is substantially smaller that the reciprocal of the distance between levels in the ladder ($\tau_p\varepsilon_E\ll 1$), the dipole moment acquires a universal shape which depends on only the temporal shape of the exciting pulse (under the condition $\tau\varepsilon_E\ll 1$, all the exponential functions can be replaced by one):

$$D(t)\propto\int_{-\infty}^{t} f(t')dt'\int_{-\infty}^{t'} f(\tau)d\tau. \tag{16}$$

The frequency of the exciting light is $\omega\approx\varepsilon_g\sim 1$ eV. If the characteristic step of the Wannier–Stark ladder is $\varepsilon_E\sim 10$ meV, the condition $\tau_p\varepsilon_E\ll 1$ holds with an ample margin at $\tau_p\approx 10$ fs. The uncertainty in the frequency of the exciting pulse itself is still small: $\tau_p\omega\sim 10$. The temporal shape of the emission signal is proportional to the second derivative of the dipole moment:[5]

$$\frac{d^2D(t)}{dt^2}\propto\frac{df(t)}{dt}\int_{-\infty}^{t} f(t')dt'+f^2(t). \tag{17}$$

It is independent of the sign of the frequency detuning, in contrast with the situation in Ref. 6.

If the pulse length is considerably larger than the reciprocal spacing of the ladder ($\tau_p\varepsilon_E\gg 1$), the exponential factors in (12) become δ-functions, and transitions occur only if the frequency is exactly equal to the difference between the energies of some pair of levels.

For an arbitrary relation between τ_p and ε_E we assume that the pulse has a Gaussian shape: $f(t)\propto\exp[-(t/\tau_p)^2]$. In this case the integrand in (12) can be expressed in terms of the error integral:[8]

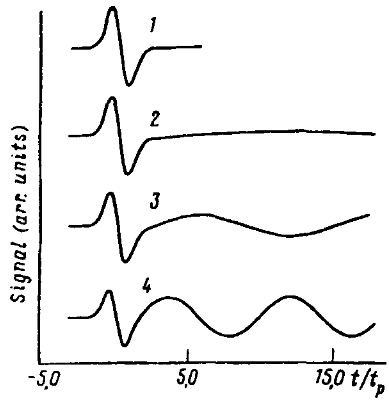

FIG. 2. Temporal shape of the emission signal. $\Delta=-\varepsilon_E$. 1–4) $\tau_p\varepsilon_E=0.1$, 0.25, 0.5, and 0.75, respectively.

$$F'(t,\omega)=\frac{\sqrt{\pi}}{4}\exp(-(\omega\tau_p)^2/4)\Phi(z), \tag{18}$$

where

$$\Phi(z)=\frac{4}{\sqrt{\pi}}\int_{-\infty}^{z}\exp(-u^2)du, \quad z=\frac{t+i(\omega\tau_p)\tau_p}{\tau_p}.$$

In the limit $t\to\infty$ the function $F(t,\omega)$ tends toward a finite limit. We then find from (15) that the dipole moment does not decay as $t\to\infty$:

$$D(t)\propto \mathrm{Re}[\exp(-i\varepsilon_E t)]$$

(in the absence of an inelastic processes and in the absence of a loss due to radiation). Strictly speaking, the dipole moment does not decay in the limit $\tau_p\varepsilon_E\ll 1$ either. During the application of the pulse, the oscillations are not manifested, since their period is greater than the time τ_p. At times $t\gg\tau_p$, they are not manifested because of inelastic processes.

Figure 2 shows the behavior of the temporal shape of the emission signal for various durations of the exciting pulse. We see that oscillations are not manifested at small values of the parameter $\varepsilon_E\tau_p$. The sign of the parameter representing the frequency detuning, Δ, does not play a fundamental role here. The shape of the signal is qualitatively the same at positive values of Δ.

The oscillations themselves are not surprising. They are analogous to the familiar oscillations in a two-level system which starts in some arbitrary state which is not an eigenstate. The decay of oscillations in the weak-field case is actually due to the quasi-continuous nature of the spectrum. In this case, transitions occur over some interval of energies. As a result, there is an integration of the oscillatory factor over this energy interval. In the simplest case we have

$$D(t)\propto\int_{\varepsilon_0}^{\varepsilon_0+eaE}d\varepsilon\cos(\varepsilon t)\propto\frac{\sin(eaEt)}{t}.$$

If the frequency lies below the threshold, the oscillatory factor is replaced by a decaying exponential factor, because of the tails on the density of states in the band gap.

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