ac Stark effects and harmonic generation in periodic potentials

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The ac Stark effect can shift initially nonresonant minibands in semiconductor superlattices into multiphoton resonances. This effect can result in strongly enhanced generation of a particular desired harmonic of the driving laser frequency, at isolated values of the amplitude.

The spectral structure of an atom can be substantially modified by an intense laser field. In particular, at sufficiently high field strengths the ac Stark effect can bring initially nonresonant levels into multiphoton resonances, resulting, e.g., in an enhancement of the photoionization signal.[1] Recent experiments[2,3] have confirmed that ac Stark effects are of central importance for a detailed understanding of the ionization dynamics.[4,5]

If we deal with a periodic lattice, instead of an atom, atomic levels broaden into energy bands. The question then arises whether a laser field can similarly be used to modify the effective band structure in a controllable way, so as to alter physical behavior. This question is of particular interest and importance for electron minibands in semiconductor superlattices,[6] where the miniband structure itself can be engineered, within wide ranges, by the design process.[7] Also, the relevant spatial size and energy scales for these structures are such that the interesting range of electromagnetic frequencies is in the far infrared, and there are now sources available which can probe them[8] with nonperturbatively strong laser fields in this spectral range. In this paper, we will show that ac Stark effects play a major role in these systems, as well. We first outline the necessary theoretical formalism, and we then demonstrate by simple numerical examples the practicality of constructing devices which make use of field-induced multiphoton resonances in superlattices for the selectively enhanced generation of particular harmonics of the driving field.

We consider a particle with effective mass m^* moving in a one-dimensional periodic potential $V(x)$ with lattice constant a. When this system interacts with an external, spatially homogeneous, electric field $\mathcal{E}(t) = \mathcal{E}_0 \sin(\omega t)$, the Hamiltonian is given by

$$\mathcal{H}(x,t) = \left[p - eA(t)\right]^2 / 2m^* + V(x) \quad , \tag{1}$$

where $A(t)$ is the electromagnetic vector potential: $\mathcal{E}(t) = -dA(t)/dt$. Because this Hamiltonian is periodic in x, the wave vector k remains a good quantum number in the presence of the external field. When transitions between different bands can be completely neglected, so that only the dynamics within a single band need be considered, then it is straightforward[9–11] to solve the time-dependent Schrödinger equation: if $\varphi_k(x) = \exp(ikx)v_k(x)$ are the Bloch-wave eigenstates for the unperturbed lattice, with energy $E(k)$, then the states

$$\psi_k(x,t) = e^{ikx} v_{q(t)}(x) \exp\left\{-i \int_0^t d\tau\, E[q(\tau)]\right\} \tag{2}$$

approximately satisfy the full time-dependent equation $i\partial_t \psi_k(x,t) = \mathcal{H}(x,t)\psi_k(x,t)$, provided that $q(t)$, which labels the periodic part of the Bloch functions $v_{q(t)}(x)$, is given by $q(t) = k - eA(t)$. Thus, this function $q(t)$ obeys[12,13] the classical equation of motion,

$$\dot{q}(t) = e\mathcal{E}(t). \tag{3}$$

This "acceleration theorem" is usually the starting point[6] for a discussion of carrier motion in semiconductor superlattices in high-frequency electric fields.

However, there is another, more general formulation of this problem. The Hamiltonian (1) is periodic not only in space, but also in time, with period $T = 2\pi/\omega$. The Floquet theorem then guarantees[14] the existence of a complete set of solutions to the time-dependent Schrödinger equation of the form

$$\psi(x,t) = \exp\left\{i\left[kx - \varepsilon(k)t\right]\right\} w_k(x,t) \quad , \tag{4}$$

with $w_k(x,t) = w_k(x+a,t) = w_k(x,t+T)$, and with "quasienergies" $\varepsilon(k)$, which specify the effect of time translation by a full period T. Thus, in the presence of the external field the energy $E(k)$ is effectively replaced by the quasienergy $\varepsilon(k)$ in characterizing the time evolution of the state labeled by quasimomentum k. Defining the Floquet functions $u_k(x,t) \equiv \exp(ikx)w_k(x,t)$, we can find the quasienergies from the eigenvalue equations

$$\left[\mathcal{H}(x,t) - i\partial_t\right] u_k(x,t) = \varepsilon(k) u_k(x,t), \tag{5}$$

with appropriate boundary conditions in space and time. Within the single band approximation quasienergies are easily determined from the wave functions (2): $v_{q(t)}$ is already periodic in space and in time, and the phase grows,

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on average, linearly with time. The quasienergy is then given[11] by the average growth rate,

$$\varepsilon(k) = \frac{1}{T}\int_0^T d\tau\, E\left[q(\tau)\right] . \tag{6}$$

For example, the standard cosine dispersion for a tight binding band of width W, $E(k) = (W/2)\cos(ka)$, yields quasienergies $\varepsilon(k) = (W/2)J_0(e\mathcal{E}_0 a/\omega)\cos(ka)$, with J_0 the zero order Bessel function.

It is crucial to recognize that the transition from the classical equation of motion (3) to the quantum eigenvalue equation (5) is not merely a reformulation. The eigenvalue equation is significantly more general; it provides a framework for attacking problems that lie outside the scope of a semiclassical treatment. For example, quasienergies remain well defined if k is no longer an exact quantum number, either because of finite lattice size or because of lattice imperfections. If the departures from spatial periodicity are local, e.g., then (5) is the starting point for an extension[15] of standard Green's function treatments of spatially local defects to systems subjected to strong time periodic fields. This reduces the problem to finite quadratures in terms of the solutions to the defect-free problem. Most importantly, the existence of dispersion relations $\varepsilon_n(k)$ (where n is a band index) relies only on the lattice spatial periodicity and the temporal periodicity of $\mathcal{E}(t)$. Therefore, these quasienergies can still be defined rigorously even when laser-induced transitions between unperturbed energy bands are no longer negligible, so that (3), and therefore (6), become invalid. Knowledge of the $\varepsilon_n(k)$ implies a nonperturbative understanding of inter(mini)band effects. It is this fact, and its consequences for possible experiments, that we will now explore in detail.

Figure 1 shows the lowest three quasienergy bands, calculated numerically, for a model potential $V(x)$ consisting of 20 square wells of width 330 Å which are separated by rectangular barriers of width 40 Å and height 0.3 eV. The effective particle mass is chosen as that of an electron in the conduction band of GaAs, $m^* = 0.067m$ (with m the bare electron mass), and the external frequency as $\omega = 3.0$ meV, as an approximate model of a $\mathrm{GaAs/Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$ superlattice in a far-infrared laser field (linearly polarized in the growth direction). If ε is a quasienergy, then so is $\varepsilon + m\omega$ for any integer m. Therefore, it suffices to consider only a single quasienergy "Brillouin zone," a range of quasienergies of width ω. The bands in Fig. 1 are labeled such that $(n, -m)$ denotes the representative of the nth quasienergy band which is shifted down in energy by $m\omega$ from that representative which is connected to the unperturbed nth energy band for vanishing laser field strength.

The first and second energy bands in the unperturbed model are separated by a gap of 12.93 meV, which is more than four times the photon energy ω. With increasing field amplitude $\mathcal{E}_0$ the bands in Fig. 1 oscillate in width and almost collapse (apart from an almost degenerate pair of edge states on top of each band) at values of $e\mathcal{E}_0 a/\omega$ equal to a zero of the Bessel function J_0. This is the behavior characteristic also of isolated bands. But, in

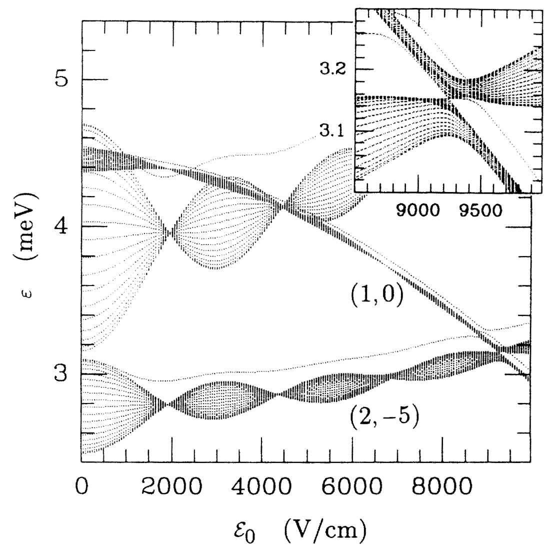

FIG. 1. Lowest three quasienergy bands for 20 square wells of width 330 Å, separated by barriers of width 40 Å and height 300 meV. The particle mass is $m^* = 0.067m$, and the ac frequency $\omega = 3.0$ meV. The inset shows a magnified view of the avoided crossing of the lowest two bands. The third band is (3,−12); it plays no role here. The split-off states are edge states (Ref. 15).

addition, interaction between different bands leads to an ac Stark shift: the lowest band (labeled $n = 1$) is shifted down when the field becomes stronger, whereas the first excited band ($n = 2$) is shifted up. Then there necessarily exists a critical field strength where the gap between the bands approaches 5ω, so that we have a field-induced five-photon resonance. In a quasienergy plot reduced to a single "Brillouin zone" such a resonance manifests itself as an avoided crossing of quasienergy bands, as shown in Fig. 1 for the representatives (1,0) and (2,−5).

At the field strength of an avoided crossing, two bands are strongly coupled. In a description based on the original energy bands one would at least have to include strong multiphoton transitions between them by higher order perturbation theory. But we can in this case achieve the nonperturbative ideal of carrying out a transformation from the original system of strongly interacting energy bands to an equivalent system of *noninteracting* quasienergy bands. The eigenvalue equation (5) already incorporates the effect of the ac field *to all orders*, and even at an avoided crossing there are no transitions at all between quasienergy states. Nevertheless, multiphoton transitions are automatically included in this picture, since the Floquet states are time-dependent linear combinations of the unperturbed band states, with maximal interband mixing at an avoided crossing.[16] Thus an analysis of the simple noninteracting description gives precise information about observable transitions in the physical system.

A field-induced avoided crossing of quasienergy bands can have observable consequences. Strong coupling between different bands, which occurs only near particular values of the amplitude $\mathcal{E}_0$, implies that the characteristics of the harmonics generated by the periodically driven lattice should alter dramatically at those values. To demonstrate this we choose an arbitrary single Floquet state $\psi(x,t)$ from the lowest band and Fourier decompose its dipole expectation value,

$$\langle\psi(t)|x|\psi(t)\rangle = \sum_n x_n \exp(-in\omega t). \qquad (7)$$

Figure 2 shows the Fourier coefficients $|x_n|$ for field strengths $\mathcal{E}_0$ below (8500 V/cm), at (9250 V/cm), and above (10 000 V/cm) the avoided crossing. Where the five-photon resonance occurs, the fifth harmonic is strongly enhanced. The response at this frequency is even stronger than it is at the fundamental frequency ω. The most general solution to the Schrödinger equation is a superposition of all Floquet states, and interferences can lead to the occurrence of additional frequencies in the dipole moment (7). However, the principal feature — strong enhancement of the mth harmonic because two bands are ac Stark shifted into an m-photon resonance — is a general result. This effect can also be found without recourse to Floquet theory by numerically analyzing the Fourier content of solutions to the Schrödinger equation at different values of the amplitude $\mathcal{E}_0$. The advantage of the Floquet method is that one does not have to search blindly for large effects as a function of $\mathcal{E}_0$, but can identify multiphoton resonances immediately from a quasienergy diagram. Thus, we now can formulate a general rule: a possible device for efficient harmonic generation by electrons in spatially periodic potentials and strong ac fields should be operated at a critical point of the quasienergy-quasimomentum dispersion relation, i.e., at an avoided crossing of quasienergy bands.

Application of this principle to semiconductor superlattices is particularly interesting, because these artificial lattices can be engineered so as to optimize the effect. Consider a simple one-dimensional tight binding Hamiltonian, which provides a good description of tunneling between different superlattice wells,

$$\mathcal{H}_{\rm TB} = E_0 \sum_\ell |\ell\rangle\langle\ell| + \frac{W}{4}\sum_\ell \left[|\ell+1\rangle\langle\ell| + |\ell\rangle\langle\ell+1|\right], \qquad (8)$$

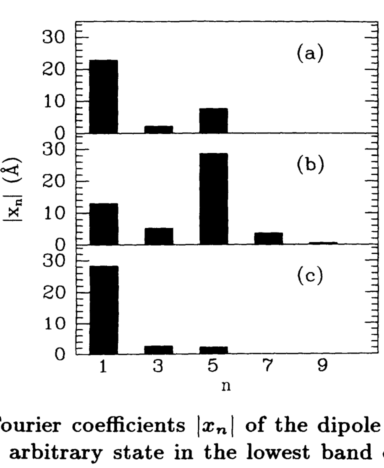

FIG. 2. Fourier coefficients $|x_n|$ of the dipole expectation value for an arbitrary state in the lowest band of Fig. 1, at field strengths $\mathcal{E}_0$ of (a) 8500 V/cm, (b) 9250 V/cm, and (c) 10 000 V/cm.

where $\{|\ell\rangle\}$ is a set of Wannier states localized in the individual wells. This Hamiltonian contains two parameters, the on-site energy E_0 and the hopping strength $W/4$. If we now start with a system described by (8), and then modify either the on-site energy (the well width) of every second site, or every second hopping integral (barrier width), then the unit cell of the periodically repeated structure consists of two wells, and the original bands each split into two. In this way it is possible to create lattices where two bands (or more, if the number of wells within a unit cell is increased further) are grouped arbitrarily close in energy, so that inter(mini)band effects play a dominant role even for small applied field amplitudes.

To illustrate this we now use a model potential $V(x)$ of 50 square wells each of width 90 Å. The separating barriers are 0.3 eV high, and their widths alternate between 40 and 60 Å. As before, the effective mass m^* is 0.067 in units of the bare electron mass. The lowest doublet for this dimerized system consists of two very narrow energy bands which are separated by a gap of 1.7 meV; this doublet is in turn separated by the much larger gap of approximately 100 meV from the first excited doublet. Figure 3 shows the quasienergy bands that originate from the lowest doublet, for an applied field of frequency $\omega = 1.0$ meV. In the two original unperturbed bands (for $\mathcal{E}_0 = 0$) corresponding states (those with the same wave vector k) are separated in the reduced zone by an energy of less than 3 meV. With increasing field strength the ac Stark effect pushes these two bands further apart, resulting first in a three-photon resonance ($3\omega = 3$ meV) and the corresponding avoided quasienergy band crossing near $\mathcal{E}_0 \approx 2200$ V/cm (see Fig. 3), and then a five-

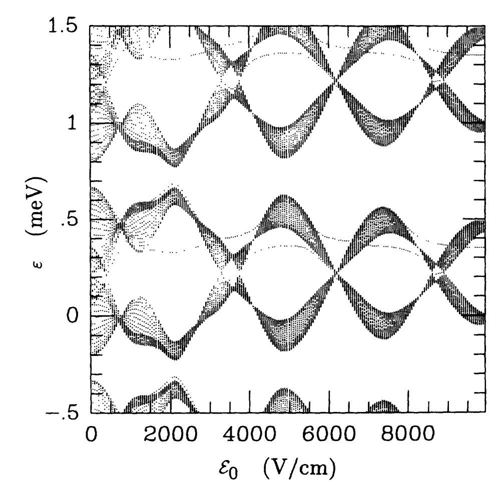

FIG. 3. Quasienergy bands from the lowest doublet, corresponding to electrons of effective mass $m^* = 0.067m$ in 50 square wells of width 90 Å, separated by square barriers of height 300 meV and widths alternating between 40 Å and 60 Å; the ac frequency is $\omega = 1.0$ meV. Two Brillouin zones are shown. Note that in the presence of the strong laser field the important dynamics are controlled not by the original energy gap of 1.7 meV, but by the much smaller quasienergy gaps of 0.1 meV (at 2200 V/cm) or 0.2 meV (at 4900 V/cm).

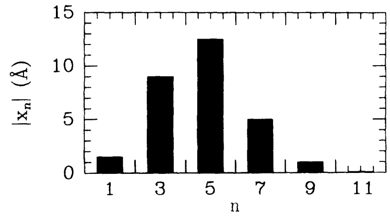

FIG. 4. Dipole expectation value Fourier coefficients for an arbitrary state at the avoided crossing near 4900 V/cm in Fig. 3.

photon resonance and the related avoided crossing near $\mathcal{E}_0 \approx 4900$ V/cm. The corresponding enhancement of the fifth harmonic generation in the latter case is exhibited in Fig. 4, which shows the Fourier amplitudes of the dipole moment corresponding to one of the states at this avoided crossing. These field strengths and superlattice parameters are well within practical experimental ranges. In general, the potential efficiency of harmonic generation associated with an avoided crossing is determined by its sharpness (the curvature of the quasienergy as a function of field amplitude at the avoided crossing; the sharper, the more efficient), which can be controlled by varying system parameters.

More than 20 years ago Tsu and Esaki[17] realized that the nonlinear optical response of conduction electrons in superlattices might find important device applications. The nonlinearity they considered explicitly was associated with the nonparabolicity of a single isolated superlattice miniband. We have shown that nonlinearities can be strongly enhanced by *inter*miniband effects which are induced by strong ac electric fields. Even in the presence of these time periodic fields the quantum behavior is simply and usefully described in terms of a few numbers — quasienergy and (for finite systems, approximate) quasimomentum. Using such a description we have demonstrated the possibility of employing the ac Stark effect to tune minibands into multiphoton resonances, and that this implies the possibility of selectively enhanced generation of a particular harmonic. The fabrication of superlattices has reached a state of great perfection,[7] and the first experiments[8] with superlattices in strong far-infrared laser fields have been successfully carried out, so it should be possible now to observe and exploit this predicted effect. More generally, the further investigation of superlattices in strong ac fields seems highly promising in terms of advances in both pure and applied physics.

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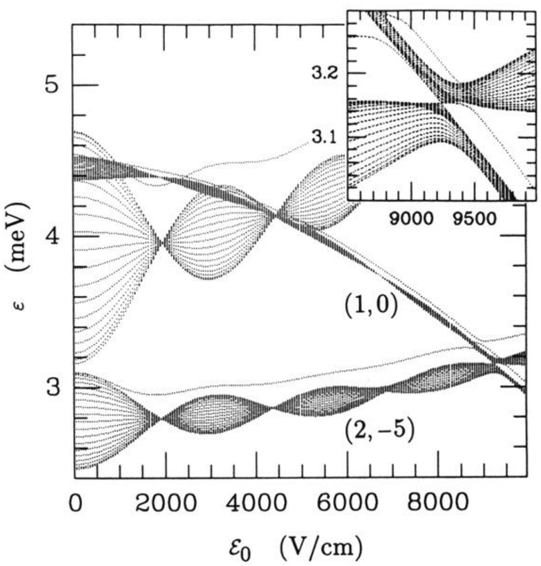

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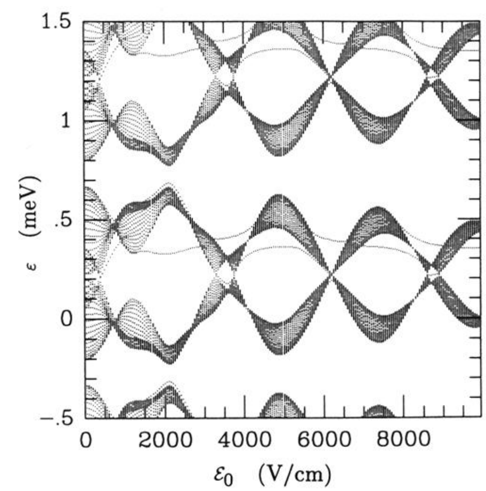

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