Strongly Driven Quantum Wells: An Analytical Solution to the Time-Dependent Schrödinger Equation

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An *analytical* solution is found for the lowest Floquet state in a single quantum well which is strongly driven by an external laser field of form $eFz\cos\omega t$. The spectral weights of the photon sidebands vary in a highly nonlinear fashion with F, exhibiting strong quenchings close to roots of the Bessel functions $J_n(k_0 eF/m\omega^2)$, where n is the sideband index and k_0 is the wave vector of the centerband resonance. The ω^{-2} scaling behavior of the roots is qualitatively different from the ω^{-1} dependence found in the coherent miniband transport in superlattices. [S0031-9007(96)00217-7]

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The recent advent of powerful free-electron lasers capable of delivering extremely strong and coherent electric driving fields has stimulated studies of photon-assisted tunneling (PAT) in nanostructured systems in the highly nonlinear regime. It may thus soon be feasible to study tailored artificial atoms and other structures subject to an intense laser field, which would provide new insight into similar experiments in atomic physics and chemistry. For strong driving, tunneling is expected to occur mainly via sidebands offset from the energy of the incident electron by a multiple of the photon energy $\hbar\omega$, with the contribution of each sideband varying nonmonotonically with the driving field [1]. Experimentally, this has indeed been qualitatively observed in superlattices [2,3] and, less conclusively, in quantum dots in the Coulomb-blockade regime [4].

There exists a good theoretical understanding of PAT in the coherent carrier transport in superlattice minibands [5,6]. Dynamical localization, leading to a collapse of the miniband and a subsequent quenching of the tunneling current, is predicted at zeros of the Bessel function $J_0(eFd_{\mathrm{SL}}/\hbar\omega)$, with eFd_{SL} the Bloch energy of the superlattice and $\hbar\omega$ the photon energy of the driving laser field. A similar quenching effect has also been predicted in biquartic double wells [7] and in potential-driven resonant tunneling diodes [8]. On the other hand, however, there is no analytical quantum-mechanical theory yet for the fundamental problem of a *single* quantum well subject to an intense laser field. In Refs. [2] and [3] the Tien-Gordon theory [1] has been used for this purpose, assuming that the applied laser field can be approximated by its potential drop across a period of the superlattice [Fig. 1(b)]. Clearly, this approximation needs critical examination.

The purpose of this Letter is to calculate the spectral function of the lowest resonance in a *single* quantum well which is strongly driven by an external laser field, and to apply the results to the analysis of *sequential* PAT in a superlattice. One of our main findings is that for this case the characteristic scale for the driving field differs from that derived by Holthaus [5] for the coherent miniband transport by a factor of the order of $E_0/\hbar\omega$, where E_0 is the energy of the quantum-well resonance. This ω^{-2} dependence of the characteristic scale is qualitatively new and cannot be explained using the Tien-Gordon theory, which predicts a ω^{-1} dependence. Also, we find an asymmetry in the photon emitting and absorbing channels which is not seen in the Tien-Gordon theory.

First, let us consider a quantum well sandwiched between two *infinitely* high walls at $z = \pm d/2$ as depicted in Fig. 1(a) which is harmonically driven by an electric field $eFz\cos\omega t$. Later we shall discuss the applicability of our theory to quantum wells surrounded by *finite* barriers. Within $-d/2 < z < d/2$ the Hamiltonian is thus of the form

$$H(t) = -\frac{\hbar^2}{2m}\frac{\partial^2}{\partial z^2} + eFz\cos\omega t\,. \tag{1}$$

A particular solution to the corresponding time-dependent Schrödinger equation was given by Truscott [9]. In order to be able to satisfy the boundary conditions one has to make an ansatz using all possible particular solutions, which for a resonant level with even symmetry yields

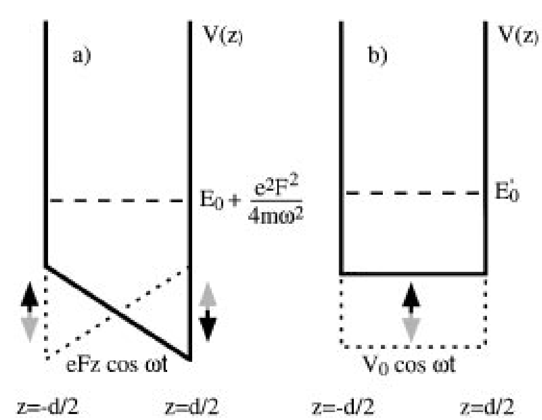

FIG. 1. A quantum well sandwiched between two infinitely high walls at $z = \pm d/2$, and driven by an external potential $eFz\cos\omega t$ generated by a laser (a) or driven by a *uniform* potential V_0 (b).

$$\psi(z,t,E_0) = \exp\left[-i\left(E_0 + \frac{e^2F^2}{4m\omega^2}\right)\frac{t}{\hbar}\right]\sum_{l=-\infty}^{\infty} A_l\left\{\exp\left[ik_l\left(z - \frac{eF\cos\omega t}{m\omega^2}\right)\right] + (-1)^l \exp\left[-ik_l\left(z - \frac{eF\cos\omega t}{m\omega^2}\right)\right]\right\}$$
$$\times \exp\left(-il\omega t - \frac{ieFz\sin\omega t}{\hbar\omega} + \frac{ie^2F^2\sin 2\omega t}{8\hbar m\omega^3}\right), \quad (2)$$

with

$$\hbar k_l = \sqrt{2m(E_0 + l\hbar\omega)}\,. \quad (3)$$

$\psi(z,t,E_0)$ is of the form $\exp(-i\epsilon t/\hbar)u(t)$ with $u(t) = u(t + 2\pi/\omega)$ which is characteristic for a Floquet state [10]. A Floquet state is the analog to a Bloch state when replacing a *spatially* periodic potential with a *time* periodic potential. The quasimomentum of the Bloch state becomes the quasieigenenergy ϵ of the Floquet state. From Eq. (2) we find $\epsilon = E_0 + e^2F^2/4m\omega^2$. The spatial symmetry of the sideband wave functions in (2) is essentially cosinelike for even sideband index l and sinelike for odd l, reflecting the fact that the applied laser field, and hence a one-photon transition from one sideband to the next, has odd parity. The energy E_0 and the coefficients A_l in Eq. (2) depend on the driving field and have to be determined from the boundary condition that the wave function vanishes at $z = \pm d/2$ at all times,

$$0 = \sum_{l=-\infty}^{\infty} A_l\left\{\exp\left[ik_l\left(z - \frac{eF\cos\omega t}{m\omega^2}\right)\right] + (-1)^l \exp\left[-ik_l\left(z - \frac{eF\cos\omega t}{m\omega^2}\right)\right]\right\} \times \exp(-il\omega t)\,.$$

To solve this equation we Fourier transform it using the identity

$$\exp\left(\frac{ik_l eF\cos\omega t}{m\omega^2}\right) = \sum_{n=-\infty}^{\infty} i^n J_n\left(\frac{k_l eF}{m\omega^2}\right)\exp(in\omega t)\,,$$

where J_n is the nth Bessel function. The boundary condition can then be recast into the form

$$0 = \sum_{l=-\infty}^{\infty}(-i)^l A_l[\exp(ik_l d/2) + (-1)^n \exp(-ik_l d/2)] \times J_{n+l}\left(\frac{k_l eF}{m\omega^2}\right) \quad \text{for all } n\,. \quad (4)$$

So far, all equations have been exact. In order to find an approximate solution to Eq. (4) we now define the dimensionless variables for the effective field strength, $q = k_0 eF/m\omega^2$, and the relative spacing of the sidebands, $v = \hbar\omega/E_0$. Note that these are implicit equations as k_0 and E_0 both depend on q and v. Expanding the wave vector $k_l = k_0\sqrt{1 + lv}$ in powers of v, and specifying further that only the lowest resonance is considered, the (not normalized) solution is found to second order as

$$A_l = i^l\left\{J_l(q) + \frac{q(q^2 - \pi^2)v^2}{64}[J_{l+1}(q) - J_{l-1}(q)] - \frac{3q^2v}{32}[J_{l+2}(q) - J_{l-2}(q)] - \frac{q^2v^2}{32}[J_{l+2}(q) + J_{l-2}(q)] - \frac{q^3v^2}{64}[J_{l+3}(q) - J_{l-3}(q)] + \frac{9q^4v^2}{2048}[J_{l+4}(q) + J_{l-4}(q)]\right\}, \quad (5)$$

with

$$k_0 d = \pi\left(1 - \frac{q^2v^2}{16}\right). \quad (6)$$

By inspecting higher-order corrections (in v) to (6) one finds that a subset of these can be summed up in a geometric series, yielding

$$k_0 d = \frac{\pi}{\sqrt{1 + q^2v^2/8}}\left[1 - \frac{(15 - \pi^2)q^2v^4}{768(1 + q^2v^2/8)^3}\right]. \quad (7)$$

The remaining unaccounted terms are of the order of $O[(v/4)^4] \times O[(qv/4)^2]$ and are generally very small unless the photon energy $\hbar\omega$ exceeds the quasieigenenergy of the resonance considerably (in which case, we have $v \gg 1$). With $E_0 = \hbar^2k_0^2/2m$ this contribution to the quasieigenenergy of the lowest driven quantum-well resonance is finally given by

$$E_0 = \frac{E_{\text{static}}}{1 + q^2v^2/8}\left[1 - \frac{(15 - \pi^2)q^2v^4}{768(1 + q^2v^2/8)^3}\right]^2, \quad (8)$$

where E_{static} denotes the energy of the resonance without any driving. Recalling that $v = \hbar\omega/E_0$, we can use the leading term of Eq. (8) to solve for v,

$$v = \frac{4E_{\text{static}}}{q^2\hbar\omega} - \sqrt{\frac{16E_{\text{static}}^2}{q^4\hbar^2\omega^2} - \frac{8}{q^2}}\,. \quad (9)$$

This equation yields real solutions only up to a maximal value $q_{\max} = \sqrt{2}\,E_{\text{static}}/\hbar\omega$, beyond which the theory breaks down. At this point, we have $v_{\max} = 2\hbar\omega/E_{\text{static}}$, giving $q_{\max}v_{\max} = 2\sqrt{2}$ and $E_{0\min} = E_{\text{static}}/2$. In terms of the field strength F one finds with Eq. (7) that the breakdown occurs at $deF/\hbar\omega = \pi$, i.e., when the ac voltage drop across the quantum well exceeds the photon energy by a factor of π. The origin of this breakdown can be traced back to the expansion of the wave vectors as $k_l = k_0\sqrt{1 + l v}$ which fails for k_l imaginary and k_0, v real, which is precisely what happens for sidebands below the bottom of the quantum well. This is also reflected by the fact that $q_{\max}$ is proportional to $E_{\text{static}}/\hbar\omega$. We stress, however, that numerical solutions of Eq. (4) do not suffer from this breakdown.

Resonant tunneling experiments on double-barrier structures have shown that the presence of a resonance in the quantum well strongly affects the transmission probability and hence the $I(V)$ characteristics of a device. Much information on the transmission probability can be drawn from the analysis of the spectral function of the quantum-well resonance, which is defined as the imaginary part of the retarded Green's function and describes the probability distribution over the sidebands n. In the case of a laser-field driven quantum well, this function is found to be $S(E) = \sum_n w_n |A_n|^2 \delta(E - E_0 - e^2F^2/4m\omega^2 - n\hbar\omega)/\sum_n w_n|A_n|^2$, where the weight resulting from the spatial integration is evaluated to be $w_n = |1 + (-1)^n J_0(2k_n eF/m\omega^2)\sin(k_n d)/k_n d|$. From Eq. (5) one sees that the spectral weights of the sidebands vary dramatically with the amplitude F of the applied ac field, and a characteristic feature found in all sidebands is that their weights go to zero at particular values of F, which for $v \ll 1$ are determined by roots of $J_n(k_0eF/m\omega^2)$. This is qualitatively very similar to what has been found in potential-driven [*à la* Fig. 1(b)] resonant tunneling diodes [8]. In what follows, we shall use this quenching effect to study how well the analysis of the spectral function of a quantum well having *infinitely* high walls can predict the transmission characteristics of double-barrier diodes with *finite* barrier heights.

Based on a numerical implementation of the transfer-matrix method to solve for the scattering states in an ac field driven double-barrier diode, we find the ac field at which the tunneling current through the centerband $n = 0$ first quenches as a function of the photon energy. Figure 2 shows the results for two double-barrier structures having $V_b = 300$ meV (solid circles) and $V_b = 700$ meV (solid squares), with $d_b = d_{\text{qw}} = 5$ nm. This has to be compared with a determination of $F_{\min}$ from the collapse of the spectral weight of the centerband using Eq. (5), which to lowest order in v gives $F_{\min} = q_0 m\omega^2/k_0 e$, where $q_0 \approx 2.4048$ is the first root of J_0 (dashed lines). For photon energies larger than the tunneling linewidth Δ of the static resonance, indicated as vertical dashed lines in Fig. 2, the analytical solution is in perfect agreement with the transfer-matrix calculation. At $\Delta \approx \hbar\omega$ a crossover is seen to a new type of ac response which, it turns out, shows a much less pronounced quenching effect in the tunneling current. This latter regime is obviously not covered by the analytical solution (5) as the assumption of infinitely high walls implies having already taken the limit $\Delta \to 0$. In passing, we note that for large values of $v = \hbar\omega/E_0$, we see from Eq. (5) that the quenching is not related in a simple way to roots of Bessel functions anymore, and hence we do not expect a simple ω^2 dependence. Nevertheless, judging from Fig. 2 the deviation appears to be rather small.

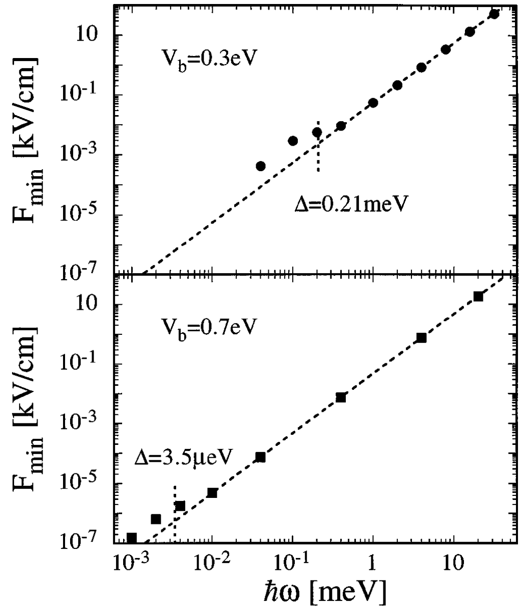

FIG. 2. Driving field strength as a function of laser frequency at which the centerband of the lowest quantum-well resonance collapses for two double barrier structures having different barrier heights. Discrete points are taken from transmission calculations, dashed lines from roots of Eq. (5).

It is interesting to compare these results with the theory of dynamical localization due to PAT in the miniband of a superlattice as discussed by Holthaus and co-workers [5], where the characteristic driving scale $eFd_{\text{SL}}/\hbar\omega$ shows a ω^{-1} rather than a ω^{-2} dependence. With Eq. (7) one finds that

$$q = \frac{k_0 eF}{m\omega^2} \approx \frac{2}{\pi}\frac{E_0}{\hbar\omega}\frac{eFd}{\hbar\omega}, \qquad (10)$$

and hence that in the present case of a *single* quantum well the characteristic scale for the ac driving field is a factor $\gamma \approx (2/\pi)(E_0/\hbar\omega)d/d_{\text{SL}}$ larger than the scale found by Holthaus. The reason behind this difference is that, while in a single quantum well the relevant driving force is indeed the ac *electric field*, in a coherently coupled two-well system or a superlattice with a coherence range longer than the superlattice period the relevant driving force is, to leading order, the ac *potential drop* between two neighboring quantum wells. Which scaling factor is more appropriate is thus entirely a question of the coherence length in the system and can be checked experimentally by varying the ratio $\hbar\omega/E_{\text{static}}$ by either changing the laser frequency or the well parameters.

We can now apply our theory to the experiment by Keay *et al.* [2], where a free-electron laser was used to study sequential PAT in a superlattice. Because of the sequential nature of the tunneling, it is sufficient to consider a single quantum well. In Fig. 3 we show the spectral function of a quantum well of width $d = 16$ nm [11], corresponding to $E_{\mathrm{static}} = 21.923$ meV, as a function of the driving laser field strength F. The photon energy was taken from experiment to be $\hbar\omega = 5.38$ meV, and hence $v = \hbar\omega/E_0 \approx 1/4$ is rather large. The top panel of Fig. 3 displays a numerical solution of Eq. (4), while the bottom one is based on an analytical fourth-order solution (in v) of the same equation. The agreement between these two is excellent up to field strengths of the order of $F \approx 4.5$ kV/cm, where sidebands below the bottom of the quantum well start to become important. The weights of the sidebands vary greatly as a function of the driving field, with the channels involving the *emission* of photons ($n < 0$) being generally stronger than those involving the *absorption* of photons ($n > 0$). This is in contrast to the theory by Tien and Gordon [1], which predicts the spectral weights of the $\pm n$ sidebands to be equal, and it may help to explain the asymmetry of these channels found in the experiment leading to absolute negative conductivity. From Eq. (5) we see that this asymmetry gets stronger with increasing ratio $v = \hbar\omega/E_0$. The authors of Ref. [2] estimate the ac field strength of their free-electron laser by fitting the minima and maxima of the sideband weights deduced from experiment to the Tien-Gordon theory. Using our theory, we find that this estimate should be reduced by a factor $\gamma \approx 2$. Clearly, for smaller values of $\hbar\omega/E_0$ the discrepancy will become even larger.

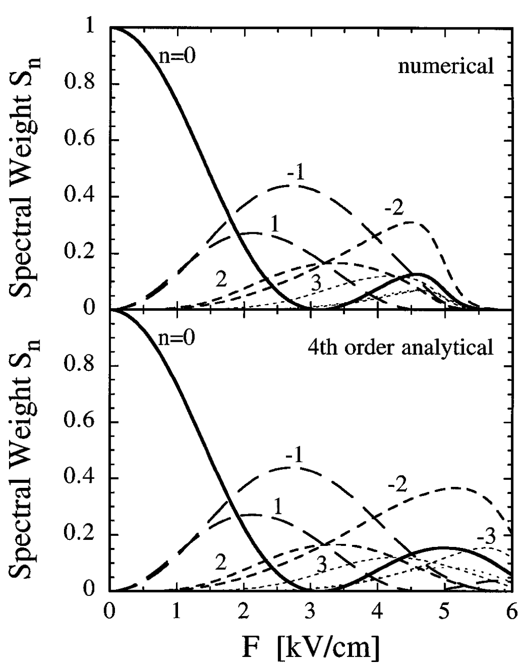

FIG. 3. Sideband spectrum of a driven quantum well of width $d = 16$ nm as a function of a driving laser field of strength F and photon energy $\hbar\omega = 5.38$ meV. The top panel is a numerical solution of Eq. (4), the lower one an analytical fourth-order solution (in $v = \hbar\omega/E_0$) in the spirit of Eq. (5).

The difficulty with transport measurements lies in the fact that the $I(V)$ and $I(\omega)$ characteristics do not directly measure the spectral function, but rather its convolution with—at least—the supply function in the emitter of the device, and/or the spectral function in the neighboring quantum wells. On the other hand, optical methods such as absorption measurements are capable of providing spectral information with very high energy resolution. We therefore propose an induced absorption experiment on a single quantum well utilizing two lasers, where a strong FIR laser is used to drive the states in the quantum well, while a second, tunable, low-power laser measures the absorption. In this way, it should be possible to trace the spectral weights and energies of a number of sidebands as a function of the power of the FIR laser.

In conclusion, we have presented an analytical and numerical treatment of a single quantum well *strongly* driven by an external laser field $eFz\cos\omega t$. In such a field a static quantum-well resonance becomes a Floquet state consisting of a series of sidebands n at energies $E_0 + e^2F^2/4m\omega^2 + n\hbar\omega$. The sideband amplitudes are a highly nonmonotonic function of the driving field F and scale with $k_0 eF/m\omega^2$, where k_0 is the centerband wave vector of the resonance. This is in contrast to the scaling law found for potential-driven quantum wells, or superlattices, where a ω^{-1} dependence was found.

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