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On non-linear optical properties of semiconductor heterostructures

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Abstract. It is shown that Floquet states follow adiabatic changes of laser pulses unless quasi-energies meet avoided crossings. At these avoided crossings quantum systems jump from one Floquet state to another with significant probability rates. These transitions, which we call the optical Landau–Zener transitions, lead to modifications of the power spectrum of scattered radiation, increasing its background significantly. It is shown, on the basis of numerical model calculations, that there is a connection between the chaotic behaviour of scattered radiation, measured by the autocorrelation function of its power spectrum, and the spectrum of quasi-energies for different light intensities. Namely, it appears that the more avoided crossings a quantum system passes during the switching on and off of the laser pulse the more chaotic is the power spectrum of scattered radiation. Resonant scattering of electrons by quantum wells is also considered.

1. Introduction

Recently the non-linear optical processes in semiconductor heterostructures (and, to a lesser degree, metal crystallites embedded in transparent dielectrics) have received considerable attention. This interest is motivated and justified on both fundamental and technological grounds. The technological aspect is related to the need for materials the optical properties of which can surpass those of electronic devices, or can open new possibilities in information processing and transmission that are not accessible with present day electronics technology (Flytzanis and Oudar 1986, Hutcheson 1987). The current attention is mostly directed towards artificial heterogeneous semiconductor microstructures where quantum confinement plays a central role and the full understanding of which is challenging for fundamental research. In one dimension such a confinement has been extensively studied in quantum wells, because their fabrication techniques have reached a high degree of precision and sophistication (see, e.g., Kelly and Weisbuch 1986). In contrast, the study of confinement effects in three dimensions has not kept up the same pace because of the lack of good fabrication techniques, these are still rather primitive when compared with those used to make one-dimensional quantum wells and other artificial microstructures (see, e.g., Kastner 1993, Beaumont and Torres 1990).

The semiconductor nanostructures occupy a position intermediate between a molecule and the bulk semiconductor. Therefore, it is a delicate problem to decide which model should be chosen in order to describe as well as possible the physical properties of such materials. Certainly, such a choice is dictated by the prominence of one feature over another but also by the complexity of the underlying calculations. For materials interacting with strong electromagnetic radiation the situation is much worse, because virtually no theoretical methods developed for radiationless problems can be adapted for this case. The reason of

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this is that strong radiation not only significantly modifies physical processes, such as for instance high-harmonic generation, but also crucially modifies matter itself to such an extent that its properties cannot further be explained in terms of radiationless quantities or notions, as will be shown shortly.

Recently we have observed increasing interest in optical intersubband processes in semiconductor heterostructures such as for instance quantum wells. This interest is due, in part, to the large dipole matrix elements for these transitions (West and Egalsh 1985, Mii *et al* 1990, Asai and Kawamura 1991). Among others, such processes as infrared detection (Levine *et al* 1989), second-harmonic generation (Rosencher *et al* 1989, Sirtori *et al* 1991), and non-linear refraction and absorption (Cotter *et al* 1991, Morrison and Jaros 1990a, Walrod *et al* 1991, Burt 1993b) have been investigated. Moreover, it has been found that for a deep and wide quantum well the dipole matrix element for an interband transition can be much larger than that for allowed intersubband transitions (Burt 1993a). This means that studies of non-linear optical properties of semiconductor heterostructures in the presence of intense laser fields become more and more important not only from the fundamental point of view, but also from the applied one. We discuss in this paper some qualitative aspects of the interaction of intense radiation with matter, without relating them to a particular quantum system.

Since we are interested in the interaction of matter with radiation generated by lasers, i.e., with a special kind of radiation characterized by its very high intensity and its coherence properties, the quasi-classical approximation of a laser field becomes adequate (see, e.g., Manakov *et al* 1986). For a single-mode field this approach consists in treating the electromagnetic vector potential not as an operator but as a function that fulfils the classical Maxwell equations. For a multi-mode field, however, such an approximation is not further applicable because the quantum character of radiation implies that in the limit of large intensities the electromagnetic vector potential has to be treated as a stochastic process, an ensemble average over which should be performed at the end (Białynicki-Birula and Białynicka-Birula 1976, Mittleman 1982). This means that it suffices to determine different kinds of cross section for a single-mode laser field and afterwards to average them over stochastic changes of laser-field parameters. This paper deals with the first part of this procedure. The second part is usually much more difficult to perform, especially if one wants to account for the interaction of radiation with matter in a non-perturbative manner; for a low-frequency radiation field a general procedure was presented by Kamiński (1988). One can read more about the quasi-classical approximation for the interaction of an intense radiation field with matter in the articles by Białynicki-Birula and Białynicka-Birula (1976), Manakov *et al* (1986) and Ehlotzky (1985), and the book by Mittleman (1982). Let me also note that the quasi-classical and the quantum formalisms in the limit of large intensities give equivalent results and that the notion '*multiphoton processes*' can also be used in the quasi-classical formalism.

The plan of this paper is as follows. In sections 2 and 3 we consider an n-state quantum system interacting with the laser pulse. The motivation for considering this model is the fact that most of the physical systems we must deal with fall into the category of 'unsolvable' quantum systems; that is, the solution of the corresponding equations of motion cannot be expressed in a form of integrals and special functions the properties of which are very well known (this is, I would say, a 'nineteenth century' definition of solubility), or the dynamic equations cannot be solved numerically quickly, so that it is not possible to analyse the response of a system to the 'almost continuous' change of parameters that describe the quantum system itself or determine the coupling to the external field. In our thinking about such complicated systems it is useful to have as a guide a simple system that can be treated quickly and in detail. That is what the present work is intended to provide. The n-state model has been frequently used in condensed-matter physics. For instance, it has been analysed in the context of optical bistability in solids (Goll and Haken 1980, Gibbs 1985) or in the study of ultrafast optical response in semiconductor heterostructures (Morrison and Jaros 1990b, Morrison *et al* 1989, Kamiński 1991). Of course such a model is greatly oversimplified because, for instance, it neglects the relaxation processes. However, it is hoped that for ultrafast optical pulses considered in this work these processes can be neglected and the predictions obtained are qualitatively correct. It is shown in section 2 how excited states of quantum wells or conducting subbands of superlattices can efficiently be populated by a short laser pulse. It appears that this phenomenon can be linked to the so-called optical Landau–Zener transitions between Floquet states at the avoided crossings of quasi-energies (Kamiński 1991). In section 3 we show that non-adiabatic Landau–Zener transitions lead to a strong enhancement of the power spectrum of scattered laser pulses and to the strong generation of higher harmonics. In particular we observe additional peaks in the power spectra that are strictly related to these non-adiabatic transitions. In section 4 we analyse a two-well model showing that 'random' distribution of avoided crossings of quasi-energies and the chaotic scattering of laser pulses are closely connected with each other. Section 5 contains the discussion of the resonant scattering of electrons by quantum wells. It is shown that resonant peaks in the reflection probabilities are strongly shifted when the intensity of radiation increases. In our investigations we have neglected the position dependence of the effective mass†, which could lead to some exotic phenomena (see, e.g., Lèvy-Leblond 1992). In section 6 we present some suggestions for further investigations.

2. n-state quantum system in strong laser pulses

The dynamics of an n-state quantum system interacting with a strong laser pulse is governed by the Hamiltonian

$$H(t) = \sum_{i=1}^{n} E_i |i\rangle\langle i| + \lambda(t)\sin(\omega t) \sum_{i,j=1}^{n} d_{ij} |i\rangle\langle j| \qquad (2.1)$$

where $\lambda(t)$ is an envelope of the oscillating electric field, slowly changing in time (as compared to the period of oscillations of electric field, i.e., to $T = 2\pi/\omega$), which is supposed to describe the laser pulse, E_i are the energy levels without the external field, and d_{ij} are the electric dipole transition moments between these levels. Such a model can easily be studied numerically (even with the help of personal computers if n is not too large) because it reduces to the finite system of ordinary differential equations, for which there exist accurate numerical subroutines (see, e.g., Shampine and Gordon 1975, Hairer *et al* 1987). From the very beginning of quantum mechanics an n-state quantum system has served as its paradigm‡, i.e., as a sufficiently simple conceptual model that embodies the important features of a large class of problems that can be considered by quantum mechanics. Despite the obvious limitations of such models they provide systems that can be studied in detail and can give a faster and deeper insight for more realistic quantum systems.

† The effective mass approximation for semiconductor heterostructures is analysed in the articles by Burt (1992) and Bastard *et al* (1991).

‡ The crucial role of this notion in physics is discussed by Kuhn (1962, 1977).

Since even such a simple model is difficult to study with the help of purely analytical methods if we want to investigate non-perturbative effects, we shall concentrate on numerical analysis of this model and on the graphical presentation of results. Let us consider first the three-level system characterized by its energies $E_1 = 0$, $E_2 = 1.25$ and $E_3 = 1.27$, and by the non-vanishing electric dipole moments $d_{12} = 2$, $d_{13} = 1$†. Moreover, the laser field is determined by its central frequency $\omega = 0.3$ and by the form of its envelope, which we choose as‡

$$\lambda(t) = \begin{cases} \lambda \exp(-t^2/2\sigma_1^2) & t < 0 \\ \lambda \exp(-t^2/2\sigma_2^2) & t > 0 \end{cases} \tag{2.2}$$

where $\sigma_1 < \sigma_2$. The asymmetric form of this envelope is to describe a real laser pulse, which is usually rapidly switched on and then slowly switched off.

Having defined all parameters of our system we can solve numerically the Schrödinger equation and calculate the probabilities $p_i(t)$ that the system at time t is in the unperturbed state $|i\rangle$. For sufficiently intense laser fields these probabilities are very rapidly changing functions of time. Therefore, in order to avoid 'black areas' in figures, these probabilities are plotted for a sufficiently dense set of discrete values of t that for times for which $p_i(t)$ change their values slowly (this corresponds to weak electric fields) the plots look like continuous curves, as in fact they are. We see in figures 1 and 2 that the first excited state is finally not populated although for intermediate times the probability $p_2(t)$ significantly differs from zero. Hence, we observe the selective population only of the second excited state $|3\rangle$§. Such a 'strange' behaviour can be understood by looking at the spectrum of quasi-energies for our model.

Quasi-energies for systems driven by a force periodic in time are the straightforward generalization of energies of stationary states (see, e.g., Manakov *et al* 1986). That is, for an envelope of the electric field constant in time, i.e., for $\lambda(t) = \lambda$, the Hamiltonian of our system is a periodic operator of time with the period equal to $T = 2\pi/\omega$. Thus, due to the Floquet theorem, an arbitrary solution of the Schrödinger equation is a linear combination of Floquet states that fulfil the following conditions:

$$\begin{aligned} |\psi_i(t)\rangle &= \exp(-\mathrm{i}E_i(\lambda)t)|\phi(t)\rangle \\ |\phi_i(t+T)\rangle &= |\phi_i(t)\rangle \qquad \text{for } i = 1, \ldots, n. \end{aligned} \tag{2.3}$$

The quantities $E_i(\lambda)$ are called quasi-energies. Quasi-energies are defined modulo ω, which means that the spectrum of quasi-energies consists of an enumerate number of the so-called Brillouin zones of width equal to ω. We encounter a similar situation in solid-state physics where the quasi-momentum is also not defined uniquely.

† Such a physical system can be realized in practice by an asymmetric quantum well (Mii *et al* 1990). In our further investigations we consider systems with other non-vanishing dipole elements arriving at the same qualitative results. This shows that the phenomenon discussed in this paper is a generic one; i.e., independent of a chosen quantum system.

‡ Let me emphasize that the numerical values of parameters of our three-level system are not crucial for the analysis presented in this paper; one could equally choose other values arriving at the same qualitative results (see the article by Kamiński (1991) where the discussion of units is also presented).

§ Let me note here that many chemical reactions can occur much faster or take place at all provided that at least one of the reactants is in a given excited state (Whitehead 1988). As we have seen such a selective excitation can be performed with a short laser pulse. This means that the laser pulse itself plays the role of let us say *'an unconventional catalyst or enzyme'*. Other aspects of controlling quantum dynamics with laser pulses are discussed in the article by Warren *et al* (1993).

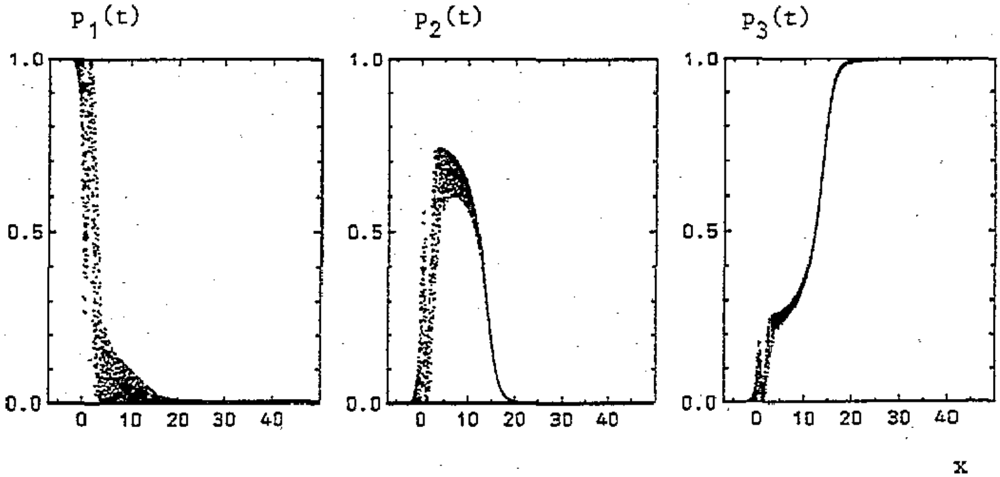

Figure 1. Occupation probabilities $p_i(t)$ for $i = 1, 2, 3$ as functions of the scaled time $x = t/10T$, $T = 2\pi/\omega$, $\omega = 0.3$ (in arbitrary units). The laser pulse is determined by equation (2.2) with $\sigma_1 = 10T$, $\sigma_2 = 100T$ and by the peak intensity defined by the dimensionless parameter $\lambda_{sc} = \lambda\omega^{-3/2} = 1.63$. The three-state quantum system is characterized by its energies $E_1 = 0$, $E_2 = 1.25$, $E_3 = 1.27$ and by the electric dipole elements $d_{12} = 2$, $d_{13} = 1$ (in arbitrary units). Let us emphasize that the occupation probabilities $p_i(t)$ are continuous functions of time. However, they change their numerical values very rapidly for times corresponding to 'sufficiently' large electric fields. Therefore, in order to avoid 'black areas', these functions are plotted for 'sufficiently' dense discrete values of t that in domains where the probabilities do not change their values rapidly the plot looks like a continuous curve (as in fact it is). The ground state is initially occupied with the probability equal to unity.

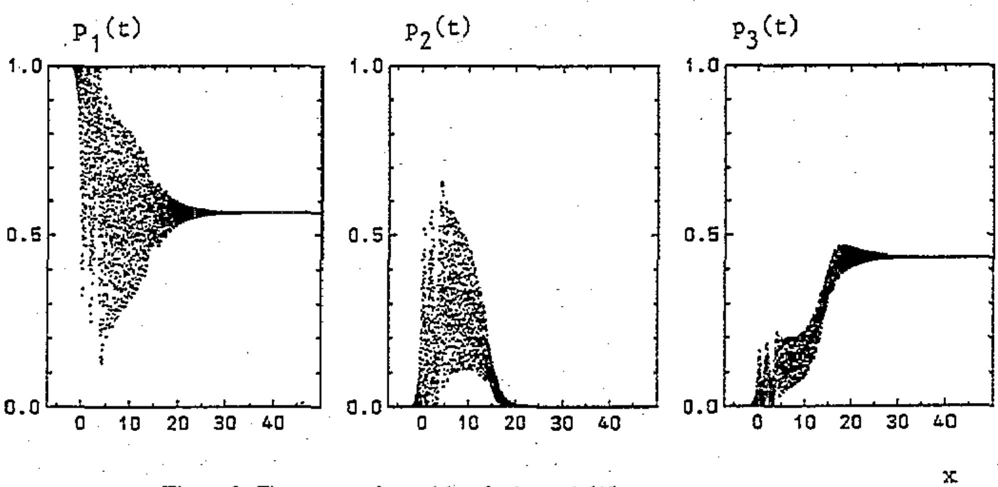

Figure 2. The same as figure 1 but for $\lambda_{sc} = 1.645$.

Two Brillouin zones of the spectrum of quasi-energies as functions of the dimensionless parameter $\lambda_{sc} = \lambda\omega^{-3/2}$ (for which units $\hbar = c = m = 1$ are used here) for our three-level model are plotted in figure 3. This figure exhibits the avoided crossing of quasi-energies

$E_1(\lambda)$ and $E_3(\lambda)$ for $\lambda_{sc} < 2$, whereas the first avoided crossing for $E_2(\lambda)$ appears for $\lambda_{sc} > 2$. This difference in the behaviour of quasi-energies is precisely the reason that only the third state is excited for $\lambda_{sc} = 2$. The reason why the avoided quasi-energy crossings are so important for the dynamics of quantum systems interacting with laser pulses is that in the vicinity of an avoided crossing the corresponding Floquet states change their form significantly. This means that for sufficiently short pulses, they do not have enough time to accommodate to a new 'environment' and the system jumps from one Floquet state to another with a significant probability. Quantitatively, the probability P of such a jump is described by the formula (Breuer and Holthaus 1989)†

$$P = \exp\left(-\frac{\pi}{2}\frac{\delta\epsilon\,\delta\lambda}{\dot{\lambda}}\right)\bigg|_{\lambda=\lambda_c} \tag{2.4}$$

where $\delta\lambda$ is equal to a change of the intensity of electric field for which the distance between the corresponding quasi-energy levels within a given Brillouin zone increases by the factor $\sqrt{2}$, whereas $\delta\epsilon$ is the distance between these quasi-energies at the avoided crossing. $\dot{\lambda}$ is the time derivative of $\lambda(t)$ and λ_c is the position of the considered avoided crossing. It follows from this expression that system does not jump from one quasi-energy surface to another provided that

$$\left(\frac{\delta\epsilon\,\delta\lambda}{\dot{\lambda}}\right)\bigg|_{\lambda=\lambda_{sc}} \gg 1. \tag{2.5}$$

Indeed, the numerical calculation shows that for our three-level model and for $\lambda_{sc} = 2$ the state $|3\rangle$ is excited with a negligible probability if the times of switching on and of switching off are very long compared to the period of oscillations $T = 2\pi/\omega$, i.e., for very small $\dot{\lambda}$ and given $\delta\epsilon$ and $\delta\lambda$. This fact is also confirmed by the avoided crossing of two excited states at approximately $\lambda_{sc} = 0.5$. This avoided crossing is so broad that it does not effect the final population of the first excited state $|2\rangle$.

It follows from figures 1 and 2 that even a relatively small increase of the peak intensity of a laser pulse (in our case from $\lambda_{sc} = 1.63$ to $\lambda_{sc} = 1.645$) can lead to a significant change of population probabilities of excited states. In fact these probabilities are oscillating functions of the peak intensity. Such oscillations have been known since 1932 in atomic collisions and are called the *Stueckelberg oscillations* (Stueckelberg 1932). They depend on the form of avoided crossings and will be discussed elsewhere.

3. Avoided crossings and scattering of laser pulses

The behaviour of a system in the vicinity of avoided crossings has a significant impact on the time dependence of induced electric dipole moments, which leads to modifications of the power spectrum of the observed scattered light. In order to show this let us consider a two-level model, the quasi-energy spectrum of which is plotted in figure 4. It follows from this figure that the first avoided crossing appears at λ_{sc} just greater than two. We can expect, therefore, that for $\lambda_{sc} < 2$ the power spectrum of scattered light will consist

† Let me note that this is the quantitative expression for the transition probability only for very sharp avoided crossings and remains a qualitative one for broad avoided crossings; a mathematically rigorous discussion of the so-called 'exponential approach to the adiabatic limit' is presented in the articles by Hagedorn (1991), Joye and Pfister (1991) and Jakšić and Segert (1992).

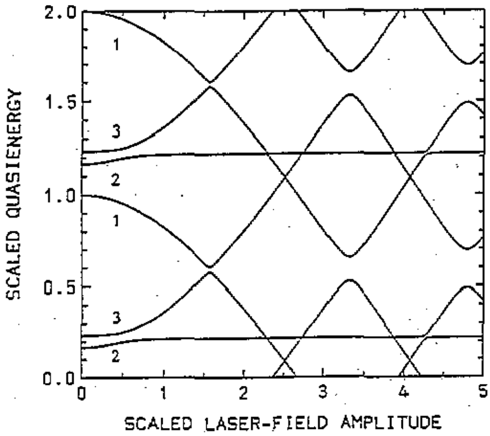

Figure 3. Two Brillouin zones of the Floquet spectrum (quasi-energies $E_i(\lambda)$ modulo ω) for the three-state quantum system characterized by its energies $E_1 = 0, E_2 = 1.25, E_3 = 1.27$ and by the electric dipole elements $d_{12} = 2, d_{13} = 1$ (in arbitrary units). The quantities $E_i(\lambda)/\omega$ are plotted as functions of dimensionless parameter $\lambda_{sc} = \lambda\omega^{-3/2}$ for $\omega = 0.3$ (in arbitrary units). The numbers on the curves show which quasi-energy curves correspond to the unperturbed energies E_i.

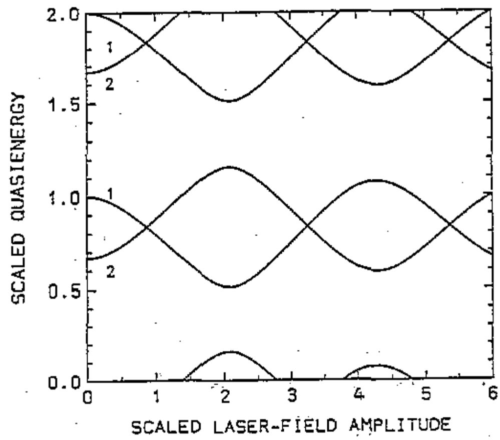

Figure 4. The same as figure 3 but for the two-level model with $E_1 = 0$, $E_2 = 1$, $d_{12} = 1$ and $\omega = 0.6$.

of separate peaks with frequencies that are multiples of the central frequency of the laser pulse. On the other hand, for $\lambda_{sc} \gtrsim 2$ we should observe qualitatively new effects. Indeed, such a situation takes place, as is illustrated in figure 5. In this figure in the upper row are plotted power spectra of scattered light for the Gauss envelope (2.2) but for different peak intensities. In the lower row are plotted power spectra for the same peak intensity but for different shapes of the pulse, chosen as:

$$\lambda(t) = \lambda \frac{2\exp(t/\sigma_1)}{1+\exp(t/\sigma_1 + t/\sigma_2)} \tag{3.1}$$

and

$$\lambda(t) = \begin{cases} \lambda/\cosh(t/\sigma_1) & t < 0 \\ \lambda/\cosh(t/\sigma_2) & t > 0 \end{cases} \tag{3.2}$$

The non-perturbative method of calculation of power spectra of scattered radiation has already been presented in the articles by Eberly *et al* (1989) and Potvliege and Shakeshaft (1989). However, for the sake of clarity we describe the main steps of this method, which in principle consists in numerical integration of the Schrödinger equation with the Hamilton operator (2.1). This equation consists of the system of n ordinary differential equations, which we write as

$$\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}a_i(t) = E_i a_i(t) + \lambda(t)\sin(\omega t)\sum_{j=1}^{n} d_{ij}a_j(t) \tag{3.3}$$

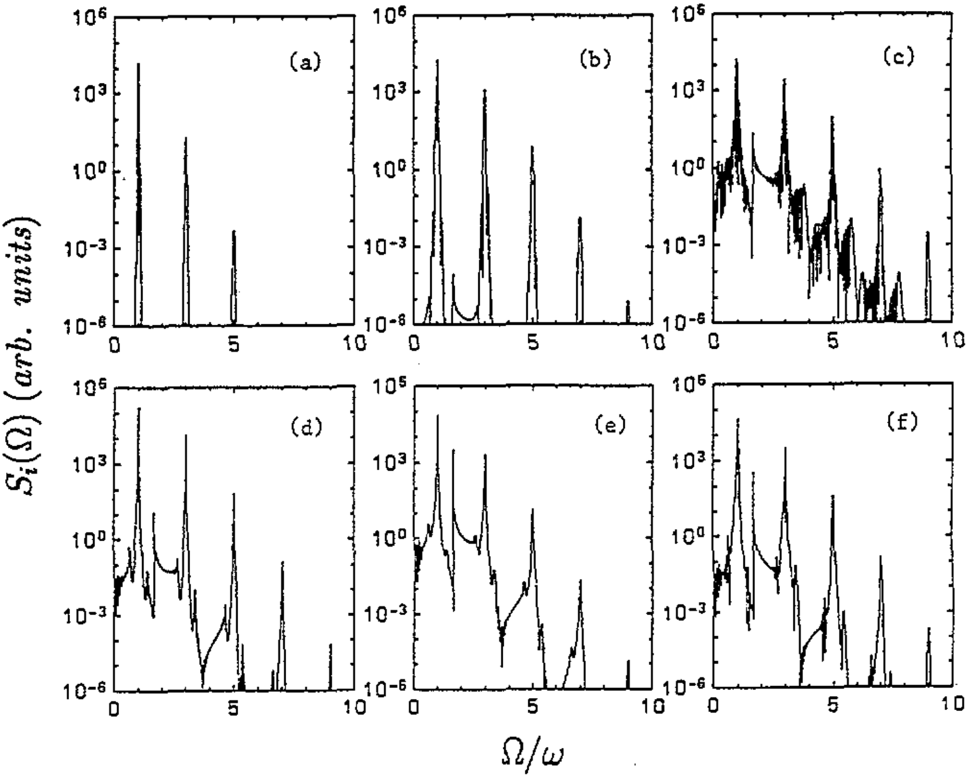

Figure 5. The power spectra $S_i(\Omega)$ for $i = 1$ as a function of Ω/ω for $\omega = 0.6$ and for the two-state model characterized by its energies $E_1 = 0$, $E_2 = 1$ and by the electric dipole element $d_{12} = 1$. The ground state is initially occupied. In the upper row the laser pulse is defined by the envelope (2.2) with (a) $\lambda_{sc} = 1$, $\sigma_1 = \sigma_2 = 10$ (in the laser period units); (b) $\lambda_{sc} = 2$, $\sigma_1 = \sigma_2 = 10$; (c) $\lambda_{sc} = 3$, $\sigma_1 = \sigma_2 = 10$. In the lower row $\lambda_{sc} = 2$ with (d) $\sigma_1 = 10$, $\sigma_2 = 50$ and the envelope (2.2); (e) $\sigma_1 = 5$, $\sigma_2 = 20$ and the envelope (3.1); (f) $\sigma_1 = 5$, $\sigma_2 = 20$ and the envelope (3.2).

in which the time-dependent function $a_i(t)$ is equal to the probability amplitude of detecting the unperturbed state $|i\rangle$ in the quantum state $|\psi(t)\rangle$,

$$|\psi(t)\rangle = \sum_{i=1}^{n} a_i(t)|i\rangle. \tag{3.4}$$

Knowing this solution we can calculate the time-dependent dipole element $d(t)$,

$$d(t) = \sum_{i,j=1}^{n} a_i^*(t) d_{ij} a_j(t) \tag{3.5}$$

and the power spectrum of scattered radiation $S(\Omega)$, which is proportional to the modulus squared of the Fourier transform of $d(t)$†,

$$S(\Omega) \sim |\, \Omega^2 \int_{-\infty}^{\infty} \mathrm{d}t\, \mathrm{e}^{-\mathrm{i}\Omega t} d(t)|^2. \tag{3.6}$$

† This is in fact the well known result from classical electrodynamics, which states that the power spectrum of light radiated by a time-dependent electric dipole is proportional to the square of the Fourier transform of the second time derivative of the electric dipole moment (see, e.g., Jackson 1975).

Let me emphasize here that for an n-state quantum system studied in this paper we have (not accounting for statistical mixtures of states) n different power spectra $S_i(\Omega)$; $S_i(\Omega)$ corresponds to the case in which in the remote past our quantum system has been prepared in the ith pure state. In our further considerations we assume that in the remote past the system was in its ground state.

One can learn from figure 5, that the power spectrum becomes more and more complicated as the peak intensity increases and that for $\lambda_{sc} \gtrsim 2$ there appears an additional peak, the position of which is independent of the peak intensity of a laser pulse. Thus, any avoided crossing leads to the appearance of an additional peak and to the increase of the background in the power spectrum. The physical interpretation of this fact is as follows. At the avoided quasi-energy level crossing (let us say of i and j levels) the Floquet wavefunction, which corresponds before the avoided crossing to the ith quasi-energy, splits after the avoided crossing into the superposition of two Floquet wavefunctions. This splitting can symbolically be written as

$$|\psi_i(t)\rangle \rightarrow a(t)|\psi_i(t)\rangle + b(t)|\psi_j(t)\rangle. \tag{3.7}$$

The probability of passing from $|\psi_i(t)\rangle$ to $|\psi_j(t)\rangle$ is given by formula (2.4). Consequently, such splittings will induce changes in time of the electric dipole moment during the switching on and off of the laser pulse. Hence, the power spectrum of scattered light will be modified. Such modifications are caused by the interference of two Floquet states in the above formula, but also by changes of forms of $|\psi_i(t)\rangle$ and $|\psi_j(t)\rangle$ at the avoided crossing. This means that one can expect a new structure in the power spectrum provided that the coefficients $a(t)$ and $b(t)$ differ significantly from zero or unity, or in other words, the avoided crossing is not too sharp (in this case $a(t) \simeq 0$) or too broad (in this case $b(t) \simeq 0$). Positions of additional peaks caused by avoided crossings can depend on the form of the laser pulse. Since in our investigations the laser pulse has a long (in time) plateau for zero intensity (i.e., when the laser pulse is slowly switched on and off), the additional peak in figure 5 corresponds to the transitions between Floquet states at zero intensities. For laser pulses that have plateaus at non-zero intensities we can observe additional peaks located at different positions. This aspect will be addressed elsewhere.

To recapitulate, this result suggests that for a given peak intensity of a laser pulse, the more avoided crossings appear in the spectrum of quasi-energies the more complicated, if not to say *chaotic*, the power spectrum of scattered light is observed. Such a relation indeed holds as we are going to show shortly.

4. Two-well potential model and scattering of laser pulses

Quantum chaos, as far as we know, was introduced in the 1970s as a quantum counterpart of the well defined classical chaos (Schuster 1984, Eckhardt 1988, Izrailev 1990, Nettel 1992). However, after many years of very intense investigations quantum chaos is still not clearly defined. Moreover, it seems that classical chaos is suppressed or at least strongly inhibited by quantization, which means that chaos is probably absent in our microscopic world and that fluctuations observed in quantum systems would have no relations with classical deterministic chaos. Examples in which quantum chaos could appear are for instance atomic systems in the presence of a strong magnetic field (Friedrich and Wintgen 1989, Hasegawa *et al* 1989) and Rydberg atoms interacting with a microwave field (Casati *et al* 1987). Moreover, it has been shown that in classically chaotic systems the quantum uncertainty, in spite of being assumed to be extremely small so as to make the classical approximation possible, increases very quickly, which makes these classical systems strongly quantum (Bonci *et al* 1993, Barone *et al* 1993). Such macroscopic quantum systems cannot be isolated from the environment because even the weakest interaction with the external world produces non-negligible effects. Hence, the search for a macroscopic manifestation of quantum chaos leads to studying the effects of environmental fluctuations on quantum systems, as was proposed for instance by Caldeira and Leggett (1983). We shall not deal with these problems here.

On the other hand results presented in section 2 suggest that at sufficiently intense laser pulses the electromagnetic radiation is chaotically scattered by matter. Indeed, for weak laser pulses frequencies of the scattered light are multiples of the frequency of the incident light. However, for strong pulses the situation becomes much more complicated, because as well as peaks corresponding to the previously mentioned frequencies, there also appears a very strong background in the power spectrum. This means that we observe a 'random' scattering of light, which could lead to the definition of quantum chaos for systems interacting with a strong laser pulse.

For realistic condensed-matter systems the situation is much more complicated than for the two- or three-level models considered so far. The number of avoided crossings that a given quasi-energy surface passes during the switching on and the switching off of laser pulses can be very large. A detailed numerical analysis of such a complicated system is impossible without the use of supercomputers, but even in this case one has to make far-reaching simplifications. For this reason we shall deal further with a still simple, although sufficiently interesting, two-well model defined by the following potential:

$$V(x) = \begin{cases} -V_0 & |x| < a \\ 0 & a \leqslant |x| \leqslant b \\ +\infty & |x| > b. \end{cases} \tag{4.1}$$

In our further analysis we shall fix the parameters of the laser pulse, i.e., the shape of the envelope, peak intensity ($\lambda_{sc} = 6$) and the central frequency ($\omega = 2$), and we shall change the parameter b for given V_0 and a. The spectra of quasi-energies for three increasing values of b are presented in figures 6, 7 and 8. We see that this spectrum becomes more and more complicated as the 'control' parameter b increases. For $b = 1$ the ground state does not meet any avoided crossing for $\lambda_{sc} \leqslant 6$. However, with increasing b we observe more and more avoided crossings of the quasi-energy corresponding to the ground state. Thus, we can expect that as the 'control' parameter b becomes larger and larger the power spectrum of scattered light is more and more chaotic. Indeed, such a situation is presented in figures 9, 10 and 11. As the measure of the 'randomness' of scattered light we have taken the autocorrelation function $A(t)$ of the dipole moment, defined as

$$A(t) = \int \mathrm{d}\tau \, \mathrm{d}(t-\tau)\, \mathrm{d}(\tau). \tag{4.2}$$

It appears that the amplitude of this function decreases as the 'randomness' of the power spectrum increases. The results presented in figures 9, 10 and 11 indicate the existence of classical chaos in the power spectrum of scattered light, whereas figures 6, 7 and 8 show something that could be called quantum chaos, because the existence of avoided crossings of quasi-energies is a purely quantum effect, which does not have a counterpart in classical theories, in which energy changes continuously. Thus, the chaotic scattering of realistic laser pulses (but not pulses modelled by rectangular envelopes) and the 'chaotic' distribution of avoided crossings of quasi-energies are very strongly connected with each other. We can, therefore, expect that the distribution of avoided crossings could provide a quantitative measure of quantum chaos (if one accepts this notion) for quantum systems interacting with an oscillating force. Our findings agree with suggestions already stated by Graffi *et al* (1987), Heiss and Sannino (1990), Goldberg and Schweizer (1991) and Takami (1992) where the subject of quantum chaos has directly been linked to avoided level crossings.

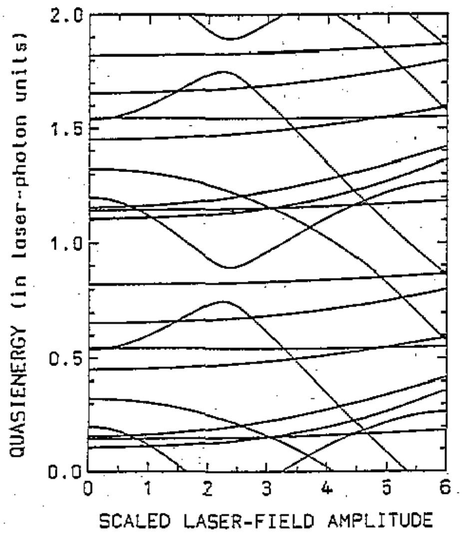

Figure 6. Two Brillouin zones of the Floquet spectrum (quasi-energies $E_l(\lambda)$ modulo ω) for the two-well potential (4.1) with $V_0 = 9$, $a = 0.2$ and $b = 1$. The quantities $E_l(\lambda)/\omega$ are plotted as functions of the dimensionless parameter $\lambda_{sc} \doteq \lambda\omega^{-3/2}$ for $\omega = 2$ (in arbitrary units). The ground-state energy $E_1 = -3.354$. We have plotted the first $N_{plot} = 10$ quasi-energies corresponding to $N_{max} = 20$ Floquet states that have the biggest projections on the first N_{plot} radiationless stationary states. For such N_{plot} the quasi-energy spectrum is stationary with respect to the increase of N_{max}. We do not plot quasi-energies of all calculated Floquet states because (i) the highest states are irrelevant (they are very weakly coupled to the ground state) and (ii) the figure would not be transparent.

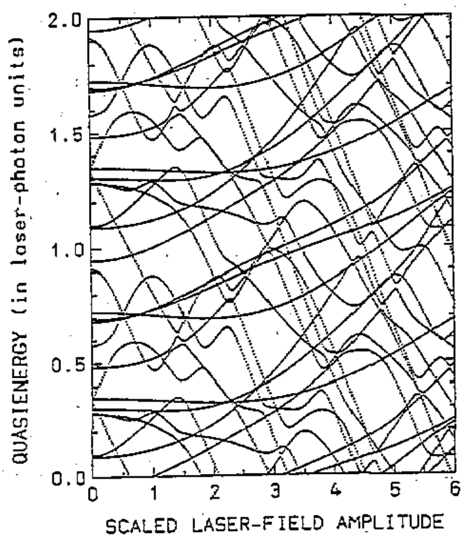

Figure 7. The same as figure 6 but with $b = 2$, $N_{plot} = 15$ and $N_{max} = 30$. The ground-state energy $E_1 = -3.442$.

5. Resonant scattering of electrons by quantum wells

Resonances are one of the most interesting phenomena in scattering processes. In the presence of the oscillating external electric field assumed to describe a laser field of a constant intensity, resonances also appear in cases when radiationless processes do not exhibit them. These resonances correspond to the possibility of an electron with energy in

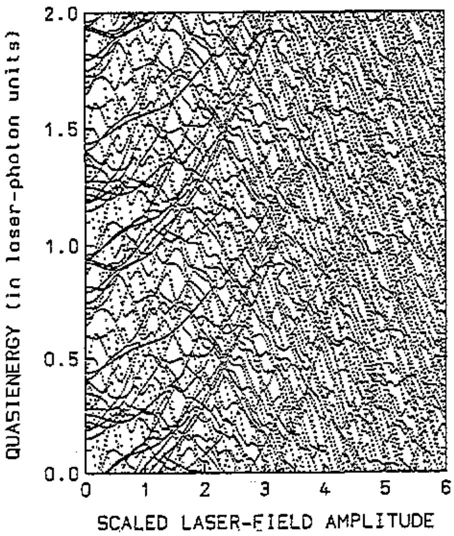

Figure 8. The same as figure 6 but with $b = 4$, $N_{\rm plot} = 30$ and $N_{\rm max} = 60$. The ground-state energy $E_1 = -3.442$.

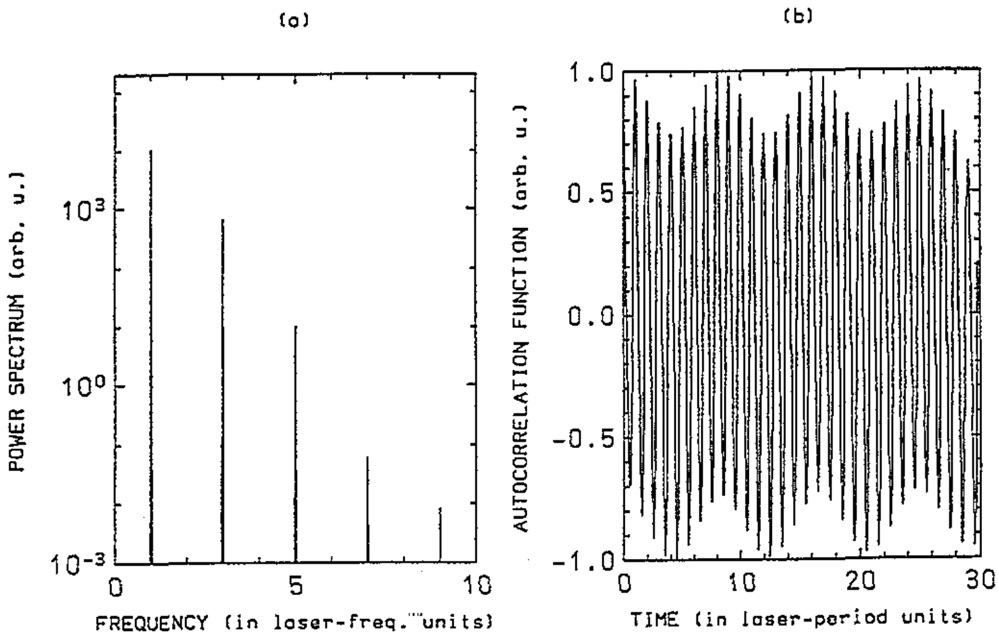

Figure 9. (a) The power spectrum $S_i(\Omega)$ for $i = 1$ as a function of Ω/ω for $\omega = 2$ and two-well potential with $V_0 = -9$, $a = 0.2$ and $b = 1$. The envelope of the laser pulse is defined by the equation (2.2) with $\sigma_1 = 50$, $\sigma_2 = 100$ (in laser-period units) and $\lambda_{\rm sc} = 6$. (b) The autocorrelation function $A(t)$ of this power spectrum as a function of t/T, $T = 2\pi/\omega$, normalized in such a way that $A(0) = 1$. Small oscillations of the amplitude of $A(t)$ are due to the fact that the ground state meets its first avoided crossing for $\lambda_{\rm sc}$ just greater than six.

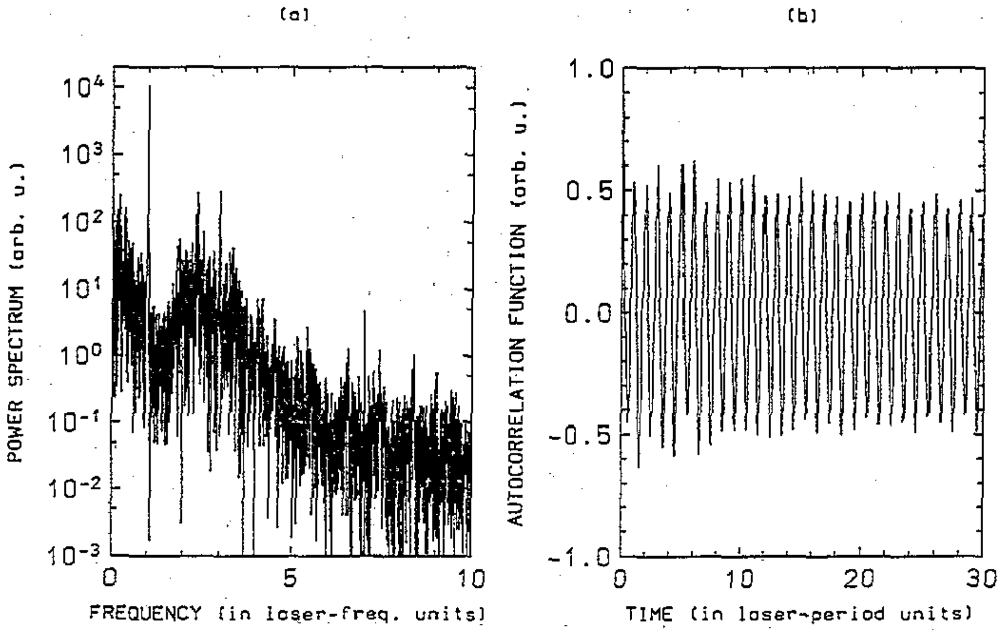

Figure 10. The same as figure 9 but with $b = 2$.

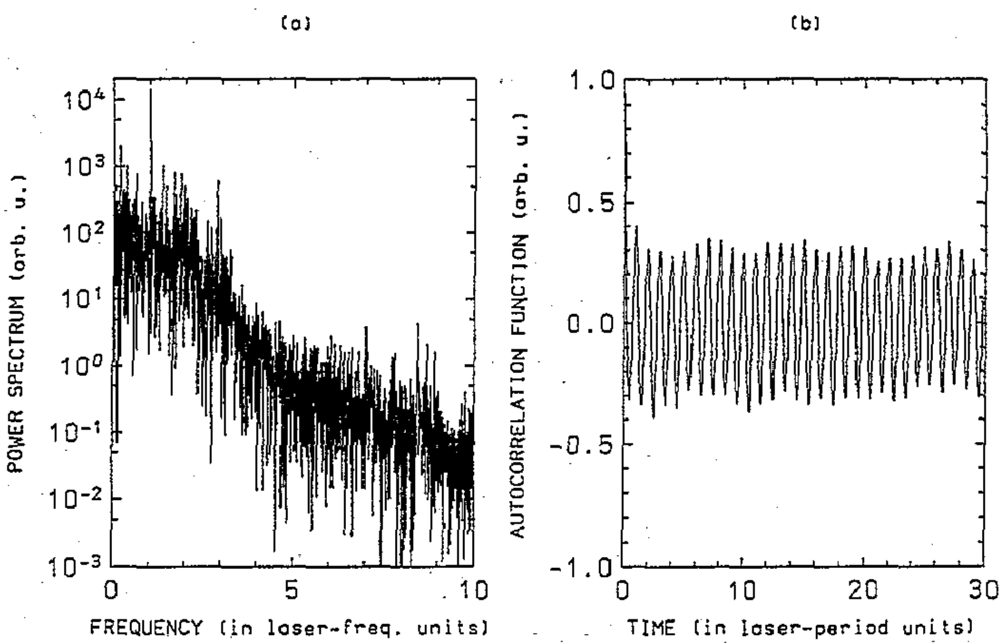

Figure 11. The same as figure 9 but with $b = 4$.

the vicinity of $E_B + k\omega$ (E_B is the energy of a bound state of the radiationless problem) emitting k photons and passing into a quasi-bound state with energy approximately equal to E_B, and then returning into an unbound state after having absorbed the corresponding number of photons. In order to study such processes we have performed an exact numerical analysis of the one-dimensional model, i.e., we have solved numerically the Schrödinger equation

$$i\partial_t\psi = \left[-\frac{1}{2m}\frac{\partial^2}{\partial x^2} + \frac{ie}{m}A(t)\frac{\partial}{\partial x} + V(x)\right]\psi \tag{5.1}$$

where the function $A(t)$ describes an electromagnetic plane wave in the dipole approximation (i.e., we neglect the space dependence of the electromagnetic phase),

$$A(t) = \mathcal{A}\cos(\omega t) \tag{5.2}$$

and $V(x)$ is a static potential intended to model a quantum well. Note that we have neglected the $A^2(t)$ term which can be eliminated by the space-independent unitary transformation, and hence does not change reflection and transition probabilities. For simplicity, the reflection and transition probabilities have been calculated for the square-well potential

$$V(x) = \begin{cases} V & |x| \leqslant a \\ 0 & |x| > a\,. \end{cases} \tag{5.3}$$

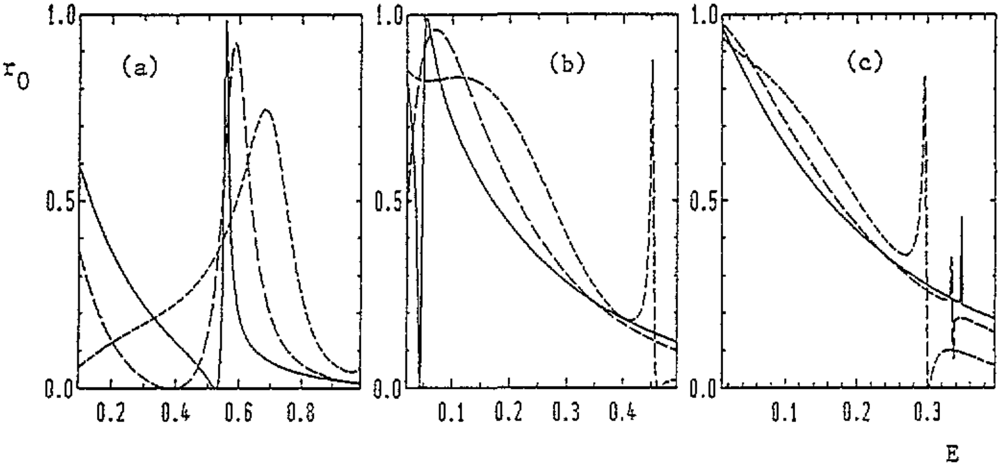

Figure 12. Elastic reflection probabilities r_0 as functions of energy for the square-well potential (5.3) with $V = -1.0$, $a = 0.7$ and for (a) $\omega = 1.0$, (b) $\omega = 0.5$, (c) $\omega = 0.4$; solid line, $\alpha_0 = 0.4$; long-dashed line, $\alpha_0 = 0.5$; short-dashed line, $\alpha_0 = 1.0$; $\alpha_0 = \mathcal{A}/\omega$ in atomic units.

In figure 12 we present the elastic reflection probabilities for the square-well potential having only one bound state of energy -0.45 (hereafter, we use atomic units). It is clear from this picture that with increasing intensity of radiation we observe a shift of the resonant peak and an increase of its width. This corresponds to the shift of the real part E_R of the complex energy $E = E_R - i\Gamma/2$ of the quasi-bound state (this is the so-called dynamic

Stark shift) and to the increase of the photoionization probability rate. It is clear from these results that the qualitative behaviour of the dynamic Stark shift depends significantly on the frequency of the laser field. For high frequencies (with respect to the bound energy) the quasi-bound states are shifted up. Such a behaviour is in agreement with theoretical predictions of the so-called high-frequency approximation (Gavrila and Kamiński 1984). On the other hand, for low frequencies of the laser field the deeply lying quasi-bound states are shifted down, which is again consistent with the results presented by Kamiński (1987, 1989, and references therein).

Let us emphasize in closing another interesting phenomenon which is shown in figure 12(b). With increasing intensity of the laser field the scattering resonance dives into 'negative energies' and appears for energies of the order of the laser photon energy. This phenomenon is due to the fact that with increasing intensity the quasi-bound state is shifted downwards to such an extent that one laser photon does not suffice to ionize the system and the electron needs to absorb an extra photon in order to jump from the discrete state to a continuous one.

6. Conclusions

In this paper we have presented some non-perturbative aspects of the interaction of semiconductor heterostructures with intense laser fields. Moreover, the laser-induced modification of the energy band structure has been considered elsewhere (Kamiński 1993). In our investigations we have assumed for simplicity that the laser field is described by a classical electromagnetic vector potential. Such an assumption is only moderately valid because the amplitude, frequency and phase of the laser field can all randomly fluctuate. This means that the real laser radiation should be described as a stochastic process. This aspect of the interaction of matter with radiation is under consideration now. Moreover, effects induced by the position-dependent effective mass are also being investigated and will be presented in due course.

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