Theory of a mixed state related to the resonance of a Stark "ladder" and optical phonons

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We study the behavior of a periodic system of quantum wells placed in a transverse electric field in the range of resonance of the frequency of an optical phonon and a Stark-transition quantum eEa, with E the electric field strength and a the lattice constant. The well-to-well transition of an electron is accompanied by emission and absorption of optical phonons of the barrier regions. In the resonance approximation the problem of a single electron interacting with phonons in a linear chain of quantum points can be solved analytically. We show that at exact resonance the electron-phonon interaction leads to delocalization of the electron. In the nonresonance case the spectrum becomes Stark-like with a quantum ω_0-eEa, with ω_0 the optical phonon frequency, and the states become localized. When the temperature is finite, the problem of states can be reduced in the adiabatic approximation to the one-dimensional strongly coupled model with off-diagonal disorder, which results in localized states even at exact resonance. We also consider the problem of the spectrum of an electron on a Stark ladder in the presence of a variable field.

INTRODUCTION

We consider a strongly coupled linear chain placed in a homogeneous constant electric field (Fig. 1). In addition, the electron interacts with either an electromagnetic field or optical phonons. We assume that the frequency of phonons (or that of the variable field) is close to the Stark quantum:

$$|eEa-\omega_0| \ll eEa,$$

where a is the lattice constant. These problems are described by the following Hamiltonians:

$$\hat{H}=\sum_n (\varepsilon_n c_n^+ c_n + V\exp\{-i\omega t\} c_{n+1}^+ c_n + V^* \exp\{i\omega t\} c_n^+ c_{n+1}), \qquad (1)$$

$$\hat{H}=\sum_n \varepsilon_n c_n^+ c_n + \sum_m \omega_0 b_m^+ b_m + \sum_n (W b_{n+1}^+ c_{n+1}^+ c_n + W^* b_{n+1} c_n^+ c_{n+1}), \qquad (2)$$

where n and m are the number of a level of the Stark ladder and the number of a localized phonon state, c_n^+ and b_m^+ the electron and phonon creation operators, and $\varepsilon_n=-eEan$ the Stark level energy ($e=-|e|$). The electric field points in the direction $-\mathbf{z}$, where $\mathbf{z}$ is the chain's axis.

A model corresponding to (1) emerges, for instance, when we consider the phototransport of electrons in a periodic lattice of plane quantum wells placed in a homogeneous electric field directed transversely to the planes. In the second problem it is assumed that the motion of electrons and phonons is limited in the plane perpendicular to the lattice's axis. The main contribution to the process of transition of an electron from well to well accompanied by emission or absorption of phonons is provided by the interaction with phonons inside the barrier. In the real situation of GaAs–AlAs-based superlattices, the phonons from the AlAs barrier are localized inside their layer, since their frequency differs considerably from that of the GaAs phonons. The transition of an electron from the $(n-1)$st well to the nth is accompanied by emission of a phonon of the nth barrier. Such processes are described by the last two terms in Hamiltonian (2). This Hamiltonian, however, cannot be employed in the case of a two-dimensional system because of the longitudinal electron momentum. In pure form, Hamiltonian (2) corresponds to a Stark ladder in a periodic chain of quantum points of the type depicted in Fig. 2.

We demonstrate that Hamiltonians (1) and (2) admit of exact solutions, localized and discrete beyond resonance and delocalized at exact resonance of the variable field or optical phonons and the Stark quantum.

The resulting coherent state of electrons and phonons can be thought of as a new type of quasiparticles in a solid, a Stark-phonon resonance, which is a mixed state of one electron and phonons, with the weak electron–phonon interaction (in the sense that $W \ll \omega_0$) at resonance ($eEa=\omega_0$) leading to the formation of a new spectrum.

In this paper we also examine more complicated states of the system, where in addition to phonons created by electrons the wells initially contained phonons. If only one excess phonon is excited, we must speak of a bound state of a Stark-phonon resonance and a phonon. This problem can easily be reduced to that of a bound state of an electron in a one-dimensional strongly coupled chain with a single defective bond that has a transition amplitude differing from the general amplitude. If phonons in all the barriers are excited, the problem is reduced to that of a strongly coupled disordered chain with off-diagonal disorder.

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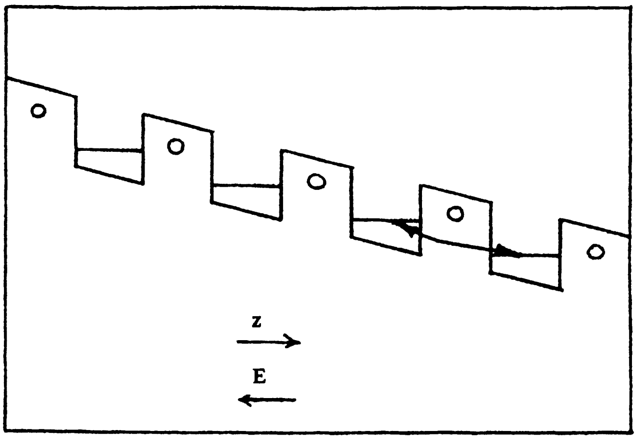

FIG. 1. The periodic potential of a sequence of quantum wells in a constant electric field. The O stand for localized phonon states.

The resonance of Stark states and optical phonons was studied earlier experimentally by Schneider, Wanger, Fujiwara, and Ploog[1] and theoretically by Bryxin and Firsov.[2] However, in contrast to the ideas of this paper, Bryxin and Firsov assumed that optical phonons enhance the dissipative process of electron drift along the electric field. In our setting the role of the resonance is to create the possibility for the existence of a collective state of one electron and optical phonons.

We limit ourselves here to the wave function of the electron–phonon system, but the results can be used to calculate the optical properties of Stark ladders. Resonance mixing between the wave function of an electron and phonons may play an important role, say, in band-to-band Raman scattering on LO-phonons.[1] In the particular case of a system with two quantum wells such a problem was studied in Ref. 3.

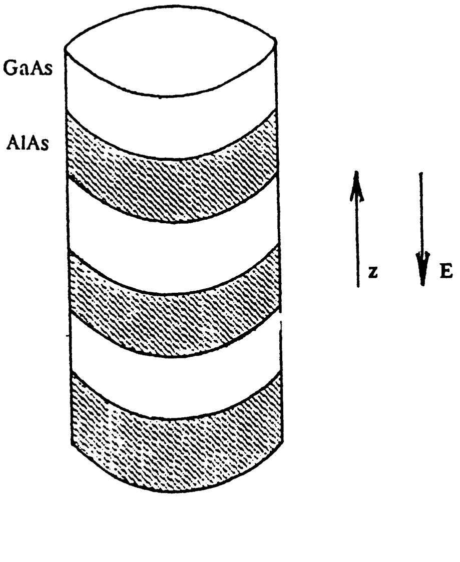

FIG. 2. A linear chain of quantum points based on the GaAs–AlAs system. Electron motion occurs in the z direction when AlAs layers interact with optical phonons.

1. THE STARK LADDER IN A VARIABLE EXTERNAL FIELD

Let us examine a one-dimensional strongly coupled chain (Fig. 1) placed in an external electric field directed along the chain's axis and consisting of a static component E and a variable component $\mathrm{Re}(F\exp\{i\omega t\})$. The system is described by the following Hamiltonian:

$$\hat{H}=\sum_n (\varepsilon_n c_n^+ c_n + V_0 c_{n+1}^+ c_n + V_0^* c_n^+ c_{n+1} + U c_{n+1}^+ c_n + U c_n^+ c_{n+1}),$$

where V_0 is the elastic tunneling amplitude, the transition amplitude $U=\mathrm{Re}(V\exp\{-i\omega t\})$ is expressed in terms of the matrix element $V=\langle\psi_{n+1}|Fd|\psi_n\rangle$ of the interaction with the electromagnetic field, with d the dipole moment, and ψ_n is the state in the number n well. If the frequency of the electromagnetic wave is close to the energy eEa of a Stark quantum, the variable field increases considerably the transition amplitude. In this situation the transition amplitude V_0 not related to the variable field can be ignored. The result is reduced to Hamiltonian (1), which defines the equation

$$\dot{c}_n=i[\hat{H},c_n] \equiv i\varepsilon_n c_n + i(V\exp\{i\omega t\}c_{n-1}+V^*\exp\{-i\omega t\}c_{n+1}). \tag{3}$$

The transformation $c_n\exp\{-in\omega t\}=a_n$ reduces the equation to

$$-i\dot{a}_n=E_n a_n + V a_{n-1} + V^* a_{n+1}, \tag{4}$$

where $E_n=n(\omega-eEa)$. Equation (4) does not contain the time explicitly. Hence, it is equivalent to the strongly coupled Hamiltonian with an overlap integral V in an electric field $E_{\mathrm{eff}}=E-\omega/ea$ but without an electromagnetic field. The solution of Eq. (4) is obvious if we employ the Fourier transformation in time, $a_n=a_n^0\exp\{i\varepsilon t\}$. However, c_n is not a stationary electron state and ε has the meaning of quasienergy rather than energy, since it is defined to within $n\omega$. Indeed, Eq. (4) is invariant under the transformation $n\to n+1$ and $\varepsilon\to\varepsilon-\omega$.

In the particular case of exact resonance, Hamiltonian (2) transforms into the periodic-lattice Hamiltonian in the tight-binding approximation. Obviously, its states are described by Bloch functions $a_n^0=\exp\{iqn\}$ with a spectrum $\varepsilon=2V\cos qa$. Out of resonance these states are Stark states, but the Stark quantum is replaced by $eEa-\omega$, that is, $\varepsilon=(eEa-\omega)n$. The states have a localization range $2V/|\omega-eEa|$. As resonance is approached, the degree of localization and the level separation decrease.

Thus, at exact resonance, Stark localized states are transformed into delocalized Bloch states. The reason is that an electron in resonance can draw from the field a limitless energy, climbing up the Stark ladder, or may emit energy in a stimulated manner, climbing down the ladder. Both processes lead to unlimited motion up or down and, respectively, to the right or left. If the frequency does not coincide with the separation of Stark levels, the resonance defect makes it possible for the electron to move virtually to a neighboring site, although the defect buildup due to multiple jumps first stops the electron and then forces it to move in the opposite direction. This localizes the state.

2. ELECTRON–PHONON STATES ON THE STARK LADDER

Here we discuss the problem of the Stark-phonon resonance spectrum for a system consisting of a linear chain of quantum points and phonon states localized in barriers between wells. Such a system can be realized by lithography and etching using the GaAs–AlAs semiconductor superlattice as a base. The system is schematically shown in Fig. 2 and consists of a "column" on the surface of the substrate. Figure 1 depicts the potential of the system in the z-direction, with the circles designating phonon states. Electron states are localized within GaAs layers (the centers of these layers have the coordinates $z=na$, with $n=0,1,2,...,N$), and phonons are localized in AlAs layers centered at points $z=(m-\frac{1}{2})a$, with $m=1,2,3,...,N$. We assume that the electron and phonon states are nondegenerate for each layer.

Let us place an electron at the level $n=0$ and consider this system near the resonance $eEa=\omega_0$. In the nearest-neighbor approximation for the electron's motion, the system Hamiltonian has the form (2), where $W=\langle\psi_{n+1}|\Phi_{n+1}|\psi_n\rangle$ is the amplitude of tunneling of the electron with emission of a phonon localized between electron quantum wells, and Φ_n the phonon potential in the respective barrier. The amplitude W is assumed to be independent of the number of a quantum point. This operator allows for the contributions of electron-phonon interaction, which are essential near the resonance $eEa=\omega_0$. Nonresonance contributions to the Hamiltonian yield only a small polaron shift in the levels of the Stark ladder of the order of $(\langle\psi_n|\Phi_n|\psi_n\rangle)^2/\omega_0$.

We construct the Stark-phonon resonance creation operator in the form

$$A^+=\sum_n \phi_n c_n^+ \prod_{m=1}^{n} b_m^+. \tag{5}$$

The unknown coefficients ϕ_n are found by solving the equation

$$[\hat{H},A^+]|0\rangle=\mathscr{E}A^+|0\rangle, \tag{6}$$

where $\mathscr{E}$ is the energy measured from the energy of state $|0\rangle$, which is a state without electrons. Actually, $|0\rangle$ may contain phonons with arbitrary occupation numbers. After commutation we obtain

$$(\varepsilon_n+n\omega_0-\mathscr{E})\phi_n+W\phi_{n-1}+W^*\phi_{n+1}(N_{n+1}+1)=0, \tag{7}$$

with N_m the occupation numbers for the state $|0\rangle$, that is, $b_m^+b_m|0\rangle = N_m|0\rangle$; these numbers can be specified arbitrarily. It is convenient to shift to new variables, $f_n = \phi_n\Pi_{m=1}^n\sqrt{N_m+1}$. This yields the equation

$$(\varepsilon_n+n\omega_0-\mathscr{E})f_n+W\sqrt{N_n+1}f_{n-1} + W^*\sqrt{N_{n+1}+1}\,f_{n+1}=0, \tag{8}$$

$$A^+|0\rangle=\sum_l \frac{f_l c_l^+\Pi_{m=1}^l b_m^+|0\rangle}{\Pi_{m=1}^l\sqrt{N_m+1}} \tag{9}$$

with the boundary conditions $f_n=0$ at $n=-1$ and $n=N+1$.

Clearly, expression (8) is the tight-binding equation for a linear chain with the amplitudes of tunneling between sites n and $n+1$ in the form $W^*\sqrt{W_{n+1}+1}$. In this section we study the case $N_m=0$, where expression (2) is an equation for the Stark level with an effective electric field $E^*=E-\omega_0/ea$ in a finite chain $0\leqslant n\leqslant N$. At the resonance $eEa=\omega_0$ Eq. (8) can easily be solved. As a result electron–phonon delocalized states emerge with the following dispersion law and amplitude:

$$\mathscr{E}(q)=2W\cos qa,\quad f_n(q)=\sqrt{\frac{2}{N}}\sin[qa(n+1)], \tag{10}$$

where N is the number of quantum wells in the chain, and $q=\pi k/(N+2)a$ the momentum ($k=1,2,...,N+1$). Obviously, in an infinitely long chain the spectrum transforms into a polaron band with a width of $4W$. The density of states in it is

$$\nu(\mathscr{E})=\frac{N}{\pi\sqrt{4W^2-\mathscr{E}^2}},\quad -2W<\mathscr{E}<2W.$$

If $eEa\neq\omega_0$, the polaron band splits into Stark levels (to each level we assign an integer p, with $0\leqslant p\leqslant N$), which correspond to localized states, with the mean coordinates $\langle\hat{z}\rangle$ of an electron in these states ($\hat{z}=a\Sigma_n nc_n$), approximately coinciding with ap. In an infinite sample Eq. (8) yields a simple expression for the energy, $p(\omega_0-eEa)$. In a finite system the wave functions are not perturbed by the boundaries if p obeys the inequalities $p\gg W/|\omega_0-eEa|$ and $N-p\gg W/|\omega_0-eEa|$ (here the quantity $\sqrt{\langle(\hat{z}-\langle\hat{z}\rangle)^2\rangle} \simeq L = aW/|\omega_0 - eEa|$ acts as the characteristic electron localization range, and the localization interval of the Stark-phonon resonance wave function f_n in the number space is $W/|\omega_0-eEa|$).

At $p\simeq W/|\omega_0-eEa|$ or $N-p\simeq W/|\omega_0-eEa|$ the position of the levels shift in relation to the equidistant position owing to edge effects, but the distance between adjacent levels is of the same order of magnitude $|\omega_0-eEa|$.

If $|eEn-\omega_0|\gg W$, the Stark-phonon resonance state consists of the localized electron state in the pth well combined with p phonons in the states m ranging from 1 to p.

We have examined the Stark-phonon resonance state near the energy $\mathscr{E}=0$. Similar states appear near $\mathscr{E}=\varepsilon_l$, where $l=1,2,...,N$. For such states to form, an electron must be placed in the lth well and released; the result is that it hops to the right with emission of phonons in the barriers. After being reflected by the boundary of the system, the electron moves to the left and absorbs the phonons emitted earlier. Thus, Stark-phonon resonance states are superpositions of Stark states with numbers $l\neq n\leqslant N$ and phonons in states $l+1 \neq m \neq N$. At resonance the energies $\mathscr{E} = \varepsilon_l + 2W \cos qa$ $(l=1,2,\ldots,N)$ can easily be obtained. The Stark-phonon resonance wave function $f_m(q)$ is nonzero for $l+1 \leqslant m \leqslant N$, and $q = \pi k/(N-l+2)a$. For an infinitely long chain the excitation spectrum consists of a number of polaron bands near the Stark energies ε_l.

When there is no resonance, the spectrum is determined by the expression $\varepsilon_l + p(\omega_0 - eEa) \simeq i\omega_0 + jeEa$, where i and j are integers. The expression corresponds to a state in which there are p phonons and the electron is centered near the $(p+l)$th well. Clearly, the expression for the energy is valid for Stark-phonon resonance states not perturbed by the boundaries. For a fixed p there are Stark-phonon resonance states with numbers p satisfying the condition $0 \leqslant p \leqslant N-l$. This shows that out of resonance each polaron band splits into $N-l+1$ Stark-like levels. In the limit of an infinitely long chain both i and j can be arbitrary. Obviously, in this case, when ω_0 and eEa are incommensurable, the states densely fill the entire real axis.

It is also worth noting that in conditions of exact resonance ($\omega_0 = eEa$) the average number of phonons in a Stark-phonon resonance state, $\langle \Sigma_m b_m^+ b_m \rangle$, is $N-p$, that is, the number of states coupled by resonance transitions that involve phonon emission.

The closely related problem of a resonance emerging in an impurity center, when the distance between two of the center's levels coincides with the optical phonon frequency, was considered by Kogan and Suris[4] (see also Levinson and Rashba's review[5]). The resonance leads to the formation of a hybrid state, with the average number of phonons in this state being roughly one. Let us compare this result with our problem. Clearly, with two quantum wells our problem is formally similar to the one considered by Kogan and Suris. But in the general case of a system of N wells the average number of phonons for a state with number p is $N-p$, since here $N-p$ electron states are in resonance.

Summarizing the ideas developed in this section, we can formulate the basic idea as follows: the electric field localizes an electron in the periodic lattice, while the interaction with optical phonons lowers the degree of localization or leads to delocalization of states in resonance. In contrast to relaxation, where the electron always moves along the field with phonon emission, in a Stark-phonon resonance state it can move in the opposite direction. Note that a Stark-phonon resonance state differs from a polaron in that in the latter the electron displaces the lattice only in its immediate vicinity, while in the former the phonons are excited at any distance from the electron.

3. A STARK-PHONON RESONANCE AT A FINITE TEMPERATURE

At a finite temperature T the number of equilibrium phonons in the sample, N_n, is finite, too. In this section we study Stark-phonon resonance states in the presence of equilibrium phonons. Let us assume that the characteristic phonon relaxation time is much longer than all quantum Stark-phonon resonance times $\hbar/\Delta E$, with ΔE the characteristic separation of Stark-phonon resonance levels. In this case the phonons in the system can be assumed "frozen." We start with Eq. (8) with the random quantities N_n having an equilibrium distribution. The probability of N_n phonons being in a given barrier is $\exp\{-N_n\omega_0/T\}$. At fairly low temperatures, $T < \omega \ln(L/a)$, where L is the Stark-phonon resonance size, this probability is lower than unity and the phonons constitute a rarefied gas.

Two approaches to this problem are possible. For low phonon number densities the states of the system can be analyzed as bound states of one, two, etc. phonons and a Stark-phonon resonance. For high phonon number densities the system can be studied via the theory of one-dimensional disordered systems.

We start with the nonresonance case. If the Stark-phonon resonance is localized between barriers containing phonons, its levels coincide with those of an unperturbed chain, $(eEa-\omega_0)n$. The states near the barriers with phonons prove to be shifted. In the limit of a strong electric field ($eEa > W$), the Stark-phonon resonance is localized at a single site. In Eq. (8) we can leave two states adjacent to the barrier that contains N phonons. The level shift is given by the second-order correction in the perturbation-theory expansion in the overlap integral:

$$\Delta = \pm W^2 \frac{\sqrt{N+1}-1}{eEa-\omega_0}.$$

The number of bound Stark-phonon resonance states is determined by the phonon distribution

$$\exp\left\{-\frac{N\omega_0+\Delta}{T}\right\}.$$

As the number of phonons increases, the nonresonance situation transforms into a devil's ladder. Obviously, this problem reduces to that of a Stark ladder in a disordered chain.

In the resonance case the theory of random disordered systems can be employed directly. We list here briefly the conclusions drawn from such an analysis. Obviously, the spectrum of the system is symmetric with respect to zero. The problem corresponds to a strongly coupled chain with off-diagonal disorder. A general statement concerning such systems is that all their states (with the exception of some) are localized and the spectrum is continuous. In view of the discreteness of the present model, the spectrum consists of separate lines (bands) corresponding to different realizations of the set of phonon numbers in the barriers (N_n).

A bound state of a Stark-phonon resonance and a phonon can be found if the phonon is localized far from the zeroth site ($m \gg 1$), by analogy with the model of an impurity state in a one-dimensional strongly coupled chain. Equation (8) assumes the form ($eEa = \omega_0$ and $l=0$)

$$\mathscr{E} f_n + W\sqrt{N_n+1} f_{n-1} + W\sqrt{N_n+1} f_{n+1} = 0,$$

$$N_n = N_m \delta_{nm}.$$

The solution is $f_n = \pm \exp\{-ka|n-m|\}$. For the energy of the bound state of a Stark-phonon resonance with a phonon at site m with N phonons we find that

$$\mathscr{E}_{\rm loc} = \pm W\left(\sqrt{N_m+1} + \frac{1}{\sqrt{N_m+1}}\right).$$

These energies lie above or below the edges of the Stark-phonon resonance band. Clearly, owing to the overlap of impurity states related to different sites, these states spread out and become an "impurity" band.

The structure in the allowed band $-2W < \mathscr{E} < 2W$ is determined by states with a wave function close to zero at points with excess phonons. This narrows the allowed band for weakly localized states to $-2W_1 < \mathscr{E} < 2W_1$, where

$$W_1 = W - \frac{\pi^2}{2ml^2},$$

with $l = a\exp\{\omega_0/T\}$ the average distance between barriers in which there are thermal excitations, and the effective mass of Stark-phonon resonance obeying the relation $m^{-1} = Wa^2$. Between these thresholds the Stark-phonon resonance states are weakly perturbed.

A special feature of a discrete system with off-diagonal disorder is that the spectrum has a singularity at $\mathscr{E} = 0$ corresponding to a weakly localized state:[6]

$$\nu(\mathscr{E}) = \frac{1}{|\mathscr{E}|\ln^3|\mathscr{E}/W|}.$$

The intervals $-2W < \mathscr{E} < -2W_1$ and $2W_1 < \mathscr{E} < 2W$ contain the exponential tails of the density of states,

$$\nu(\mathscr{E}) = \exp\left\{-\frac{\pi}{l\sqrt{m(\mathscr{E} \pm W)}}\right\}.$$

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