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1994 J. Phys.: Condens. Matter 6 4721

(http://iopscience.iop.org/0953-8984/6/25/010)

J. Phys.: Condens. Matter 6 (1994) 4721–4726. Printed in the UK

Electric-field-induced localization and non-perturbative response of a one-dimensional conductor

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Received 24 December 1993, in final form 14 March 1994

Abstract. We consider the non-perturbative non-stationary response of a one-dimensional conductor and the carrier localization induced by an external electric field within the one-electron one-band tight-binding approximation. Exact general analytical expressions for the polarization, electric current and carrier mean square displacement are obtained. We find a new effect of *absence of induced polarization* in an arbitrary electric field for an electron, initially localized on one site. If the electron wavevector change Δk during one period of the field is a multiple of 2π, the electron is typically delocalized except for the *localization* in a Bloch state *at the bottom of the band* and a *generalized dynamic localization*. If $\Delta k/2\pi$ is a non-integer, the electron remains localized. Particular cases of a harmonic field, a sum of constant and harmonic fields and of periodic pulses are also considered for which localization occurs, i.e. suppression of coherent tunnelling.

1. Introduction

Quasi-one-dimensional (Q1D) systems such as conjugated polymers exhibit interesting electric and optical properties owing to a high degree of electron delocalization along molecular chains [1–3]. Theoretical calculations of conductivity and susceptibilities typically take account of the applied field in a perturbative way up to second or third order and are limited to the pure harmonic (AC) case [1–6]. Electron dynamics in band models of crystalline conductors have been studied extensively for the case of a uniform time-independent (DC) electric field, when the spectrum transforms into a Stark ladder and Bloch oscillations occur [7–13]. A new effect called *dynamic localization* has been discovered previously by Dunlop and Kenkre (DK) [14] in a harmonic field; for certain values of the ratio E/ω of the field amplitude to its frequency, the initially strong localized electron in a one-band conductor turns out to be *localized* for arbitrary time.

In this paper we present rigorous analytical results on electron localization and a non-linear response to an electric field of arbitrary magnitude and time dependence. We consider an infinite 1D chain in the one-electron one-band tight-binding approximation with nearest-neighbour transfer integrals V in the Wannier basis set $|n\rangle$ under the action of an arbitrary electric field $E(t)$:

$$H(t) = -V \sum_n (|n\rangle\langle n+1| + |n+1\rangle\langle n|) + eE(t) \sum_n n|n\rangle\langle n|. \tag{1}$$

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0953-8984/94/254721+06$19.50

The intersite distance is unity, the electron charge is $-e$, and the position operator is assumed to be diagonal. The quantities that we are interested in are the induced polarization $\Delta d(t)$, the electric current density $j(t)$, and the electron mean square displacement $\Delta\langle n^2(t)\rangle$:

$$\Delta d(t) = -e\sum_n n\Delta\rho_{n,n}(t) \qquad j(t) = \Delta\dot{d}(t) \qquad \Delta\langle n^2(t)\rangle = \sum_n n^2\Delta\rho_{n,n}(t) \tag{2}$$

where $\Delta\rho_{n,n}(t) = \rho_{n,n}(t) - \rho_{n,n}(0)$ is the induced change in the density matrix. It is easy to see that the exact solution to the Schrödinger equation with the Hamiltonian (1) is provided by the wavefunction $|\varphi(t)\rangle = \sum_n c_n(t)|n\rangle$, where

$$c_n(t) = \exp[-\mathrm{i}n\eta(t)]\sum_r c_r(0)J_{n-r}(2|s|)\left(\mathrm{i}\frac{s}{|s|}\right)^{n-r} \qquad s = \frac{V}{\hbar}\int_0^t \mathrm{d}t'\,\exp[\mathrm{i}\eta(t')]. \tag{3}$$

J_{n-r} are Bessel functions of order $n-r$, where n, r are sites, and $\eta(t) = e\hbar^{-1}\int_0^t \mathrm{d}t'\,E(t')$ is the effective field pulse area.

2. Electron response in an arbitrary field

Rather lengthy calculations of quantities, defined in equation (2), with the use of equation (3) give the following new *exact* and most *general* solutions for the electron response [15]:

$$\Delta d(t) = 2e\rho\frac{V}{\hbar}\int_0^t \mathrm{d}t'\,\sin[\eta(t')-\kappa] \qquad \eta(t) = e\hbar^{-1}\int_0^t \mathrm{d}t'\,E(t') \tag{4}$$

$$\begin{aligned}\Delta\langle n^2(t)\rangle = 2\left(\frac{V}{\hbar}\right)^2\int_0^t \mathrm{d}t'\,\mathrm{d}t''\,\{N_\mathrm{e}\cos[\eta(t')-\eta(t'')] - \rho_2\cos[\eta(t')+\eta(t'')-\kappa_2]\}\\ - 2\rho_1\frac{V}{\hbar}\int_0^t \mathrm{d}t'\,\sin[\eta(t')-\kappa_1]\end{aligned} \tag{5}$$

where N_e is the total number of electrons in the system, and ρ_i and κ_i are determined by the non-diagonal elements of the initial density matrix:

$$\begin{aligned}\rho\exp(\mathrm{i}\kappa) &= \sum_n \rho_{n,n+1}(0)\\ \rho_1\exp(\mathrm{i}\kappa_1) &= \sum_n \rho_{n,n+1}(0)(2n+1)\\ \rho_2\exp(\mathrm{i}\kappa_2) &= \sum_n \rho_{n,n+2}(0).\end{aligned} \tag{6}$$

For an initial field-free eigenfunction (a Bloch wave) we have the following cases. Case A is

$$\rho = \rho_2 = 1 \qquad \kappa = \kappa_1 = k \qquad \rho_1 = 2\langle n(0)\rangle + 1 \qquad \kappa_2 = 2k \tag{7}$$

where k is the wavevector. Electrons in an empty band can be created, for example, by doping or photoexciting a dielectric or by injecting carriers from electrodes.

Case B is a partly filled metallic band with $\nu = 2k_F/\pi$ electrons per atom corresponding to the following substitutions (k_F is the Fermi wavevector):

$$N_e \to \frac{2N}{\pi}k_F \qquad \sin\kappa_1 \to 0$$

$$\cos\kappa \to \cos\kappa_1 \to \frac{2N}{\pi}\sin k_F \qquad \cos\kappa_2 \to \frac{N}{\pi}\sin(2k_F).$$

Case C is one or several electrons, each initially localized on one site; this case is special as it reveals a new effect, namely the *absence of induced polarization* in an arbitrary electric field. As can be seen from equations (4)–(6) $\rho_i = 0$ and $\Delta d(t) \equiv j(t) \equiv 0$; there is no induced dipole moment and no current. A non-confined electron positioned like a quasi-classical particle on one site does not shift in an arbitrary field at all!

The interpretation of this effect is the following: the coordinate Kronecker symbol δ produces a uniform filling of all the states in the band $\sim N^{-1}$ (vanishing in the thermodynamic limit). As the time evolution of electrons in an external field corresponds to the shift of the wavevectors $k(t)$ of occupied states (see, e.g., [9] and equations (4) and (5)) by the same amount $\eta(t)$, in this case the first Brillouin zone becomes shifted as a whole with time, but that does not change the matrix elements which are periodic in k. The effect is the result of coherent compensation of quantum transitions between states in the band due to periodicity in k-space.

The centre of mass of the wave packet does not move, although its width (the first term of $\Delta\langle n^2(t)\rangle$ in equation (5)) grows unboundedly. The initial delocalization of the electron destroys the effect. The effect considered is different from the cases of a fully occupied band, when there are no accessible states for the electron to make transitions to and from the *dynamic localization* [14], which we shall consider in more detail below. Although strongly localized initial conditions seem somewhat artificial, we believe that the effect can be observed at low temperatures in high-quality superlattices or quantum dot arrays; localizing an electron in a nearly macroscopic quantum well seems quite realistic.

3. Periodic fields or pulses

Next let us consider the electron localization, induced by an electric field, periodic in time (pulses included). By localization we mean the regimes when both the time dependences of the polarization and mean square displacements (4) and (5) are bounded and oscillate only, so that the average electron velocity vanishes.

For a Bloch electron (case A) equations (4) and (5) take the form

$$\Delta d(t) = 2e\frac{V}{\hbar}I(t) \qquad \Delta\langle n^2(t)\rangle = 4\left(\frac{V}{\hbar}\right)^2 [I(t)]^2 - 2\rho_1\frac{V}{\hbar}I(t) \tag{8}$$

$$I(t) = \int_0^t \mathrm{d}t'\,\sin[\eta(t') - k]. \tag{9}$$

The system evolution is determined by the change Δk in the wavevector during one period of the field.

(a) If Δk is a multiple of 2π (periodic case), i.e.

$$\Delta k = \eta(t+T) - \eta(t) = 2\pi l \qquad l \text{ integer} \tag{10}$$

then the average displacements during different periods are additive:

$$\Delta d(t=nT) = -evt \qquad \Delta(n^2(t=nT)) = (vt)^2 - \rho_l vt \qquad v = 2\frac{V}{\hbar T} I(T) \tag{11}$$

and the particle propagates with a constant average velocity v with periodic oscillations superimposed on it. As the integral over the period in equations (9) and (11) is typically non-zero, this periodic case corresponds to a delocalized regime. Two exceptions are *localization at the bottom of the band* and the *generalized dynamic localization.*

The electron in a Bloch state at the *bottom of the band* ($k = 0$) is localized if $I(t) = 0$ (equation (9)). The sufficient condition is provided by equation (10) plus the symmetry of $E(t)$ within the period

$$E(T_0 + \Delta t) = E(T_0 - \Delta t). \tag{12}$$

In fact, then the pulse area η, (equation (4)) is antisymmetric: $\eta(T_0 + \Delta t) - \eta(T_0) = -[\eta(T_0 - \Delta t) - \eta(T_0)]$ and the average velocity (equations (11) and (9)) becomes zero. All the other Bloch states ($k \neq 0$) are typically delocalized and propagate with the average velocity v, which is a multiple of the field-free value $v_0 = 2(V/\hbar)\sin k$ and the factor v_E, that characterizes the average effect of the field during one period. The latter becomes zero in the *generalized dynamic localization* regime, defined by

$$v_E \equiv T^{-1} \int_{T_0-T/2}^{T_0+T/2} \mathrm{d}t\, \cos[\eta(t)] = 0. \tag{13}$$

When equations (12) and (13) for the parameters of the electric field are satisfied, the integral $I(t)$ in equation (9), which determines the average velocity (11), becomes zero for arbitrary k. Consequently the polarization and mean square displacement (11) retain only bounded oscillating components; the electron is localized.

DK [14] calculated only the mean square displacement in a pure harmonic (AC) field for an initial state localized on one site. They found one particular form of the effect of dynamic localization but have not noted the regime of localization at the bottom of the band and could not observe the effect of absence of induced polarization as it requires the consideration of polarization and non-localized initial states. The response for an extended initial wavefunction cannot be obtained as a weighted sum of probability propagators, as the coherence of contributions originating from different sites will then be lost. The contributions of different particles are additive, while contributions from site occupation probabilities of the same particle are not; the non-diagonal elements of the density matrix should be taken into account from the very beginning. In our approach we consider the general case of arbitrary initial states.

We note that the generalized dynamic localization takes place in specific applied fields. It characterizes the spatial evolution of the wave packet but does not characterize the kinetic coefficients. In fact, the AC conductivity is determined by the oscillating component of the polarization which is non-zero, while the DC conductivity in the regime (13) should be obtained from the solution of the problem incorporating a sum of DC and AC fields. In a constant field no DC is produced, while in the low-frequency AC field the nth-order conductivity diverges as ω^{-n}, although the total AC conductivity, obtained as a sum of all order processes with the output frequency ω, does not diverge: $\sigma(\omega) = 4e(V/\hbar)\rho\cos kJ_1(\epsilon) \to 0$ as $\omega \to 0$. As we are considering a quantum problem *with time-reversible evolution without relaxation, the Nernst–Einstein formula does not* apply.

(b) The next case corresponds to an arbitrary change $\Delta k = \Delta\eta = \eta(T) - \eta(0)$ during one period, which is not a multiple of 2π. The expression for $I(t)$ in equation (9), which determines $\Delta d(t)$ and $\Delta\langle n^2(t)\rangle$ (equation (8)) is governed by

$$I(t = nT) = \frac{\sin(\frac{1}{2}n\Delta\eta)}{\sin(\frac{1}{2}\Delta\eta)}\int_0^T \mathrm{d}t\ \sin\{\eta(t) - [k - \tfrac{1}{2}(n-1)\Delta\eta]\}. \tag{14}$$

In the *commensurate* case, when $\Delta\eta/2\pi = m/m'$ is a rational number, the electron performs periodic motion with the period $m'T$ (see (8) and (14)). The particle is localized; its displacement is finite. In the particular case $m' = 2$ the electron reverses its motion after each pulse.

For $\Delta\eta/2\pi$ the *incommensurate* case, the motion is quasi-periodic. For finite time t it can be approximated with arbitrary accuracy by the commensurate case with large values of m, m'. In both cases, $\Delta d(t)$ and $\Delta\langle n^2(t)\rangle$ have no unbounded components and oscillate only, which corresponds to electron localization.

Finally, we consider some particular cases to illustrate the general classification scheme.

The pure harmonic (AC) field $E(t) = E\cos(\omega t)$ corresponds to case (a). The expression for the electron velocity, deduced from equations (8) and (9),

$$v = 2\frac{V}{\hbar}\sin kJ_0(\epsilon) \qquad \epsilon = \frac{eE}{\hbar\omega} \tag{15}$$

reveals the regimes of localization at the bottom of the band, $k = 0$ (equation (12)), and dynamic localization [14], $J(\epsilon) = 0$ (equation (13)). The zeroth-order Bessel function J_0 appears also in the field-induced renormalized spectrum of a two-level system [16].

A DC + AC field $E(t) = E_0 + E\cos(\omega t)$ belongs to classes (a) and (b), where one now has $\mu = \omega_0/\omega$ (the ratio of the Bloch frequency $\omega_0 = eE/\hbar$ to the harmonic frequency ω) an integer, a rational number or an irrational number. For μ an integer (case (i)) the electron is typically delocalized and propagates with the average velocity

$$v = 2(-1)^{\mu}\frac{V}{\hbar}\sin kJ_{\mu}(\epsilon) \qquad J_{\mu}(\epsilon) = \frac{1}{\pi}\int_0^{\pi}\mathrm{d}t\ \cos(\mu\tau - \epsilon\sin\tau) \tag{16}$$

(where $J_{\mu}(\epsilon)$ is the Anger function), except for the regimes of localization at the bottom of the band, $k = 0$, and generalized dynamic localization, $J_{\mu}(\epsilon) = 0$ (cf equations (13) and (14)). For μ a non-integer (case (ii)) the integral $I(t)$ in equation (9) is bounded:

$$I\left(t = \frac{2\pi n}{\omega}\right) = J_{\mu}(\epsilon)\frac{2\pi}{\omega}\frac{\sin(\frac{1}{2}\mu\omega t)}{\sin(\pi\mu)} \tag{17}$$

and the electron oscillates in a localized state.

Finally let us consider pulses, consisting of n periods of cosine function, repeated at time interval T. The expression for the electron velocity given by

$$v = 2V\hbar^{-1}\sin kq^{-1}[q - n + nJ_0(\epsilon)] \qquad q = (2\pi)^{-1}\omega T \tag{18}$$

consists of contributions from free-electron propagation during the part of the period $(q-n)/q$ and propagation in an AC field (15) during nT/q. Again we observe the localization at the bottom of the band, $k=0$, and the generalized dynamic localization $J_0(\epsilon)=1-q/n$.

If the pulse contains $n+\frac{1}{2}$ cosine periods, the electron is delocalized (case (a)). Its velocity in addition to (18) consists of the contribution from the propagation during half the period with different v':

$$v = 2V\hbar^{-1}q^{-1}\{\sin k[q-(n+\tfrac{1}{2})+(n+\tfrac{1}{2})J_0(\epsilon)]+\cos k\tfrac{1}{2}E_0(\epsilon)\}$$
$$E_u(\epsilon)=\frac{1}{\pi}\int_0^\pi \sin(\mu\tau-\epsilon\sin\tau)\,\mathrm{d}\tau. \tag{19}$$

Both condition (12) and condition (13) are violated; so there is no localization at the bottom of the band and no dynamic localization in this case.

In conclusion, we find that *dynamic localization* is a general phenomenon, occurring for arbitrary initial states and both DC and AC fields. It corresponds therefore to a suppression of coherent tunnelling as the degeneracy of sites is lifted temporally by the external periodic field, or by a sequence of pulses.

Acknowledgments

We thank the Natural Science and Engineering Research Council of Canada for an international exchange fellowship for KP. André Bandrauk is a member of the National Center of Excellence in Molecular Dynamics.

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