EXACT SELF-PROPAGATORS FOR QUASIPARTICLE MOTION ON A CHAIN WITH ALTERNATING SITE ENERGIES OR INTERSITE INTERACTIONS

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We present exact calculations of the amplitude and the probability self-propagators for two model systems. Both systems consist of a quasiparticle moving among the sites of an infinite linear chain via nearest-neighbour matrix elements. In one model, the site energies alternate in space but the intersite interactions are all the same. In the other model, the interactions alternate but the site energies are all the same. Similarities and differences are explored and the extreme parameter limits are studied.

In our study of quasiparticle propagation on lattices, we have recently discovered some simple but interesting solutions for the probability of an initially occupied site in two different but related one-dimensional infinite systems (chains). They are relevant to a variety of fields [1,2] including exciton motion in molecular crystals [3,4], electron transport in superlattices [5,6], and the motion of excitations in ferroelectric materials [7–9]. We report our findings below.

Both our models consist of a quasiparticle such as an electron or an excitation (e.g. a Frenkel exciton) moving among the sites of a chain via nearest-neighbour matrix elements. In one model (model A) the matrix elements are all the same and equal to V but the site energies alternate between the values $E\pm 2\Delta$. In the other model (model B) the site energies are all the same and equal to E but the nearest-neighbour matrix elements alternate between the values $V\pm\delta$. It is straightforward to calculate the band energies and to point out their similar nature in the two models. We also present two additional results which we believe are both interesting and useful for practical studies: analytic solutions of the self-propagator (amplitude as well as probability) in the time domain for both models, and the demonstration that the probability propagator envelope tends, in each model, to that in a chain with no variations of site energy and matrix element but with a new effective matrix element.

Model A. The difference in the site energies at alternate sites is taken to be 4Δ in this model and the nearest-neighbour matrix element for site-to-site transfer is V. The hamiltonian H is thus

$$H=2\Delta\sum_m(-1)^m|m\rangle\langle m| + V\sum_m(|m\rangle\langle m+1|+|m-1\rangle\langle m|)\,. \tag{1}$$

Here and elsewhere in this Letter, $|m\rangle$ represents a state localized on a lattice site m (e.g. a Wannier state), V is taken to be real for simplicity, and the summations are from $m=-\infty$ to $m=+\infty$. With the expansion of the time-dependent state $|\Psi(t)\rangle$ of the system in terms of the states $|m\rangle$ through $|\Psi(t)\rangle=\sum_m C_m(t)|m\rangle$, the evolution equation of the time-dependent amplitudes $C_m(t)$ may be obtained as

$$\mathrm{i}\partial_t C_m(t)=2\Delta(-1)^m C_m(t) + V[C_{m+1}(t)+C_{m-1}(t)]\,, \tag{2}$$

where ∂_t denotes the time derivative. Discrete Fourier transforms of the real-space amplitudes C_m give us the following equations for the Bloch-space amplitudes $C^q(t)$:

$$\mathrm{i}\partial_t C^q(t)=\Delta C^{q+\pi}(t)+2V\cos q\, C^q(t)\,, \tag{3}$$

$$\mathrm{i}\partial_t C^{q+\pi}(t) = \Delta C^q(t) - 2V\cos q\, C^{q+\pi}(t)\,. \tag{4}$$

Here q is the (dimensionless) wavevector. We see that the dispersion relation between the energy Ω_q and q consists of the two branches of

$$\Omega_q^2 = 4(\Delta^2 + V^2\cos^2 q)\,. \tag{5}$$

The explicit relationship between the amplitudes in the two spaces is

$$C_m(t) = \frac{1}{2\pi}\int_0^{2\pi} \mathrm{d}q\,\exp(-\mathrm{i}qm)\, C^q(t)\,. \tag{6}$$

The solution of the real-space amplitudes for the initial condition that the quasiparticle occupies a single site (at $m=0$) is given by

$$C^q(t) = \cos(\Omega_q t) - 2\mathrm{i}[(\Delta + V\cos q)/\Omega_q]\sin(\Omega_q t)\,. \tag{7}$$

The relevant integrals can be evaluated [10] in the Laplace domain. We concentrate on the self-propagator here. In the Laplace domain we obtain

$$\tilde{C}_0(\epsilon) = (\epsilon - 2\mathrm{i}\Delta)[(\epsilon^2 + 4\Omega^2)(\epsilon^2 + 4\Delta^2)]^{-1/2}\,, \tag{8}$$

where ϵ is the Laplace variable and $\Omega^2 = \Delta^2 + V^2$. We invert this propagator, and write for the evolution in the time domain the explicit expression

$$C_0(t) = J_0(2\Omega t) + \Phi_1(t) + \mathrm{i}\Phi_2(t)\,, \tag{9}$$

$$\Phi_1(t) = 2\Delta\int_0^t \mathrm{d}\tau\, J_0(2\Omega\tau)\, J_1(2\Delta(t-\tau))\,, \tag{10}$$

$$\Phi_2(t) = 2\Delta\int_0^t \mathrm{d}\tau\, J_0(2\Omega\tau)\, J_0(2\Delta(t-\tau))\,. \tag{11}$$

The square of the modulus of (9) gives us the probability $P_0(t)$ of the initially occupied state:

$$P_0(t) = J_0^2(2\Omega t) + \Phi_1^2(t) + \Phi_2^2(t) + 2J_0(2\Omega t)\,\Phi_1(t)\,. \tag{12}$$

In these expressions J_n is the ordinary Bessel function of order n. We see that the first term on the right-hand side of each of (9) and (12) represents the simple linear chain without the site energy variation except for the "renormalization" of the frequency,

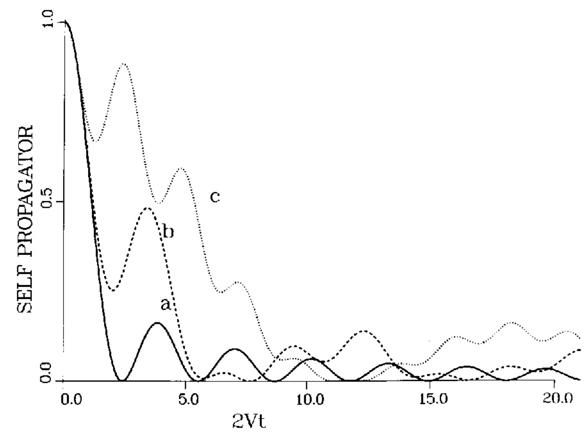

Fig. 1. The time evolution of the probability self-propagator (probability of an initially fully occupied site) in model A (see eq. (12)) for various values of the ratio Δ/V (a:0, b:0.4, c:1) showing the effect of alternating site energies on the quasiparticle propagation. A tendency to return to the initial site as a result of energy mismatch is seen clearly.

and that the quasiparticle moves slower as a result of the energy mismatch. Fig. 1 shows the probability self-propagator given by (12) for several values of the site energy variation.

Model B. Here the site energies are all equal but the nearest-neighbour matrix element for site-to-site transfer V alternates between the values V_1 and V_2. We call the difference $V_1 - V_2$ as 2δ and the average $\frac{1}{2}(V_1+V_2)$ as V. The hamiltonian H is thus

$$H = \delta\sum_m (-1)^m(|m+1\rangle\langle m| - |m\rangle\langle m-1|) + V\sum_m (|m+1\rangle\langle m| + |m\rangle\langle m-1|)\,. \tag{13}$$

The evolution equation of the time-dependent amplitudes $C_m(t)$ is

$$\mathrm{i}\partial_t C_m(t) = \delta(-1)^m[C_{m+1}(t) - C_{m-1}(t)] + V[C_{m+1}(t) + C_{m-1}(t)]\,. \tag{14}$$

The Bloch-space amplitudes $C^q(t)$ obey

$$\mathrm{i}\partial_t C^q(t) = 2\mathrm{i}\delta\sin q\, C^{q+\pi}(t) + 2V\cos q\, C^q(t)\,, \tag{15}$$

$$\mathrm{i}\partial_t C^{q+\pi}(t) = -2\mathrm{i}\delta\sin q\, C^q(t) - 2V\cos q\, C^{q+\pi}(t)\,, \tag{16}$$

and the two branches of the dispersion relation are given by

$$\Omega_q^2 = 4[\delta^2 + (V^2 - \delta^2)\cos^2 q] \,. \qquad (17)$$

For the initial condition that the quasiparticle occupies a single site initially, the real-space amplitude is given by

$$C^q(t) = \cos(\Omega_q t) + 2i[(V\cos q + i\delta \sin q)/\Omega_q]\sin(\Omega_q t) \,. \qquad (18)$$

We have also been able to evaluate these integrals in the Laplace domain. As in (7) above we concentrate on the self-propagator here. In the Laplace domain we get

$$\tilde{C}_0(\epsilon) = \epsilon[(\epsilon^2 + 4V^2)(\epsilon^2 + 4\delta^2)]^{-1/2} \,. \qquad (19)$$

An inversion yields

$$C_0(t) = J_0(2Vt) + \Phi_1(t) \,, \qquad (20)$$

$$\Phi_1(t) = 2\delta \int_0^t d\tau\, J_0(2V\tau)\, J_1(2\delta(t-\tau)) \,. \qquad (21)$$

The probability $P_0(t)$ of the initially occupied state is

$$P_0(t) = J_0^2(2Vt) + \Phi_1^2(t) + 2J_0(2Vt)\,\Phi_1(t) \,. \qquad (22)$$

We see again that the first term on the right-hand side of each of (20) and (22) represents the simple linear chain without the site energy variation. Fig. 2 shows the probability self-propagator given by (22) for several values of the transfer matrix element variation.

The slowing down of the quasiparticle apparent from figs. 1 and 2 and from eqs. (10), (12), (20), (22) for the two models is obviously expected. A more interesting behaviour is seen in figs. 3 and 4. They show the evolution of the probability self-propagator for the case when the chain disturbance is large, i.e., for $\Delta \gg V$ in model A (fig. 3) and for $2\delta^2 \gg V^2$ in model B (fig. 4). We see similarities as well as differences. Both models show that the envelope of the self-propagator follows the evolution of an *undisturbed chain (i.e. with vanishing Δ and δ)* but with an effective transfer matrix element $V_{\rm eff}$ which equals V^2/Δ for model A, and $(V^2-\delta^2)/\delta$ for model B. The dashed line in figs. 3 and 4 is $J_0^2(2V_{\rm eff}t)$, i.e. the self-propagator for the undisturbed chain. The form of $V_{\rm eff}$ shows that the disturbance (Δ and δ respectively in the two models) acts somewhat like a scattering rate: we recall [3] that a scattering rate α in an undisturbed chain with intersite matrix element $V_{\rm eff}$ gives rise to a "hopping rate" of $2V_{\rm eff}^2/\alpha$.

The difference between the two models is made apparent by the fact that wild oscillations occur under the envelope for model B but not for model A.

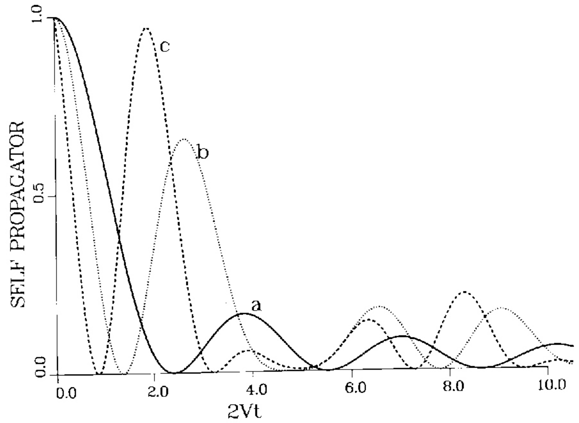

Fig. 2. The time evolution of the probability self-propagator in model B (see eq. (22)) for various values of the ratio δ/V (a:0, b:0.3, c:0.5) showing the effect of alternating intersite transfer elements on the quasiparticle propagation.

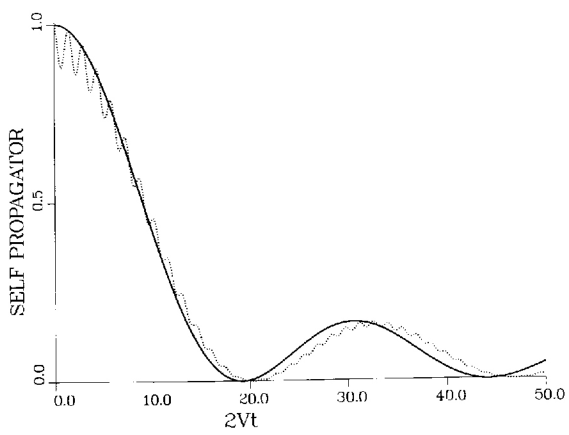

Fig. 3. The limit for large energy mismatch of the time evolution of the probability self-propagator in model A (see eq. (12)) compared to the case of no mismatch but with an effective intersite transfer. Shown is the exact evolution for model A (solid line) with $\Delta/V=2$ and an undisturbed chain result viz. $J_0^2(2V_{\rm eff}t)$ with $V_{\rm eff}=V^2/\Delta$.

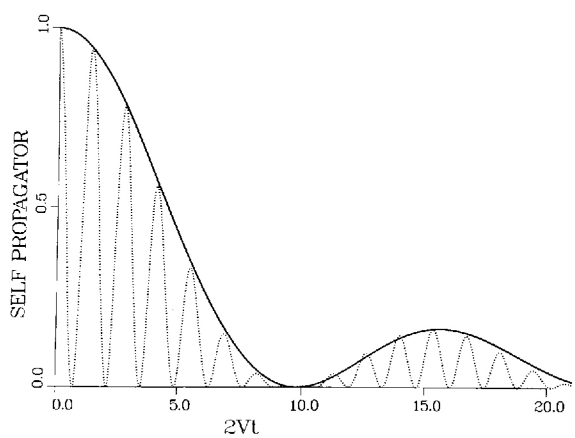

Fig. 4. The limit for large variation of the intersite matrix element (with respect to its average value) of the probability self-propagator in model B (see eq. (22)) compared to the case of no variation but with an effective intersite transfer. Shown is the exact evolution for the model B with $V_1/V_2=4$ and an undisturbed chain result viz. $J_0^2(2V_{\rm eff}t)$ with $V_{\rm eff}=\frac{4}{3}(V_1V_2)^{1/2}$.

Indeed, the limit $\Delta \gg V$ reduces the oscillations in model A but the corresponding limit $2\delta^2 \gg V^2$ enhances them in model B. This is easily understood. The oscillations describe the transfer of the quasiparticle within the two-site "unit cell" whose repetition generates chain B. The relative largeness of the matrix element V_1 which connects one site to the other in the unit cell (with respect to V_2 which connects the site to the next unit cell) causes the quasiparticle to remain largely within the cell and to move only a little from site to site. On the other hand, in model A, the quasiparticle encounters an equal energy mismatch for remaining in as well as leaving a unit cell. The sharp difference in the two models is seen as follows. When $V=\delta$ in model B, the connection between the cells is severed and the quasiparticle localized initially on the initial site oscillates (the self-propagator is simply the square of a cosine), being unable ever to leave the cell. No limit which is in obvious correspondence to this appears to be possible in model A. Indeed, it is easy to see that what does correspond to this behaviour is $V \ll \Delta$ to such an extent that V is to be neglected entirely. In such a case, the quasiparticle never leaves the initially occupied site simply because site-to-site transfer is impossible in the light of a vanishing V or enormous energy mismatch Δ. The "internal structure" naturally present in model B leads to the wild oscillation even in this "localization" limit. With the exception of this essential difference (which is representative of their being each other's "duals" in the sense of the theory of electric networks) the two models show the following correspondence in their parameters:

$$\text{model A:}\quad \Delta\,, \qquad V\,,$$
$$\text{model B:}\quad \delta=\tfrac{1}{2}(V_1-V_2)\,,\quad \sqrt{V^2-\delta^2}=\sqrt{V_1V_2}\,.$$

In conclusion, we have investigated, through exact analytical expressions for the amplitude and probability self-propagators, the motion of a quasiparticle on two infinite chains, one with alternating site energies and the other with alternating intersite transfer matrix elements. We find similarities as well as differences in the two models. The similarities are seen in the wavevector dependence of the band energies and in the limiting tendency of the self-propagators to acquire J_0^2 envelopes. The effective V's in the limiting case are V^2/Δ, $(V^2-\delta^2)/\delta$ respectively. The application of these results as well as of related extensions such as arbitrary-site propagators and correlation functions to observables in experimentally relevant situations will be reported in a future publication.

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