Extended states in disordered systems: Role of off-diagonal correlations

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We study one-dimensional systems with random diagonal disorder but off-diagonal short-range correlations imposed by structural constrains. We find that these correlations generate effective conduction channels for finite systems. At a certain golden correlation condition for the hopping amplitudes, we find an extended state for an infinite system. Our model has important implications to charge transport in DNA molecules, and a possible set of experiments in semiconductor superlattices is proposed to verify our most interesting theoretical predictions.

Electronic states in disordered system have been an active research topic for many years. Ever since the pioneering work of Anderson,[1] it has been generally believed that disorder in low-dimensional systems leads to unequivocal localization of electrons. However, the situation changes if additional structure or correlations are imposed on the statistical properties of the randomness. It was found, for example, that a few special extended states in a one-dimensional (1D) "random dimer" model exist due to symmetries of the resonant scattering in the structure.[2] Furthermore, the existence of a mobility edge separating extended and localized states was confirmed for 1D random systems with weak long-range *correlated* disorder.[3]

Most studies have concentrated on diagonal disorder, where the local energies in a tight-binding description are assigned randomly, although some studies have explored the role of off-diagonal disorder, where the intersite hopping constants are chosen from a random distribution. The role of correlated diagonal and off-diagonal disorder has received attention only recently, both theoretically[4,5] and experimentally.[6] Moreover, in many systems, local correlations appear naturally due to the built-in chemical structure. In this Brief Report, we investigate the effects of structural constraints on the correlated diagonal and off-diagonal disorder, and their impact on charge transport. We find that the local correlations generate extended states, which therefore enhance electronic transport even in the macroscopic limit.

Our studies have been motivated in part by questions on the nature of charge transport in DNA, a subject which has arisen much interest recently, due to its fundamental roles in biological processes and in possible novel device designs.[7] A DNA molecular system can be viewed as a 1D chain composed of base pairs AT and CG in a typically random order. The on-site energies for pairs of bases AT and CG are different, corresponding to the different ionization potentials.[8] The role of on-site energy correlations in different DNA sequences was discussed recently in the literature,[9] where a constant hopping amplitude was considered. However, the hopping amplitude (via π-orbital overlap) in DNA molecules depends on whether the electron (or hole) hops between AT/AT, AT/CG, or CG/CG base pairs.[10] The short-range correlation of the hopping amplitudes due to the built-in chemical structure is shown to affect the transport properties and ef-

fectively open conduction channels for electrons in DNA molecules—even in those with fully random sequences, such as λ-phage DNA. The transport properties are shown to be actually determined by a subtle competition between the disorder in base pair arrangement (on-site disorder) and hopping ("off-diagonal") correlations.

The minimal model to study random systems with diagonal and off-diagonal disorder is an effective 1D tight-binding model described by the Hamiltonian

$$H = \sum_j \left[\varepsilon_j c_j^\dagger c_j + t_{j,j+1}(c_j^\dagger c_{j+1} + c_{j+1}^\dagger c_j) \right], \quad (1)$$

where the on-site energies are chosen from the bivalued distribution $\varepsilon_j = \varepsilon_A$ and ε_B. Correspondingly, the hopping constants are given by $t_{j,j+1} = t_{AA}$ (or t_{BB}), if $\varepsilon_j = \varepsilon_{j+1} = \varepsilon_A$ (or ε_{BB}); while $t_{j,j+1} = t_{AB}$, otherwise. This model is perhaps the simplest generalization of the Anderson model, which is the limit for $t_{AA} = t_{BB} = t_{AB}$. Notice that in DNA, the *A* and *B* labels refer to the two kinds of base pairs, AT and CG, while the model could be easily adapted to describe electronic states in other complex molecules (polymers) and/or semiconductor superlattices, as we will discuss below.[11]

When the concentration of one type of site is small, say *B*, the probability for two nearby sites to have the same on-site energy ε_B is smaller. In this case, the system tends to the "repulsive binary alloy" model, in which one extended state exists.[2] A simple calculation yields the transmission coefficient for a system with one impurity with on-site energy ε_B,

$$T_1(E) = \frac{(2t_{AB}^2 \sin k)^2}{(2t_{AB}^2 \sin k)^2 + N_1^2}, \quad (2)$$

where $N_1 = W t_{AA} + 2(t_{AB}^2 - t_{AA}^2)\cos k$, $E = 2t_{AA}\cos k$, and $W = \varepsilon_B - \varepsilon_A$. One can see that for the state with energy $E = W t_{AA}^2/(t_{AA}^2 - t_{AB}^2)$, the transmission coefficient is unity. The states *near* this energy have large transmission coefficient and long localization length, even in systems with more *B* impurities. In fact, these states have an important contribution to transport. Figure 1 shows the transmission coefficient for various concentration of *B* impurities with energy ε_B. The transmission is obtained by a transfer-matrix calculation for 1000 sites, and averaged over 300 different con-
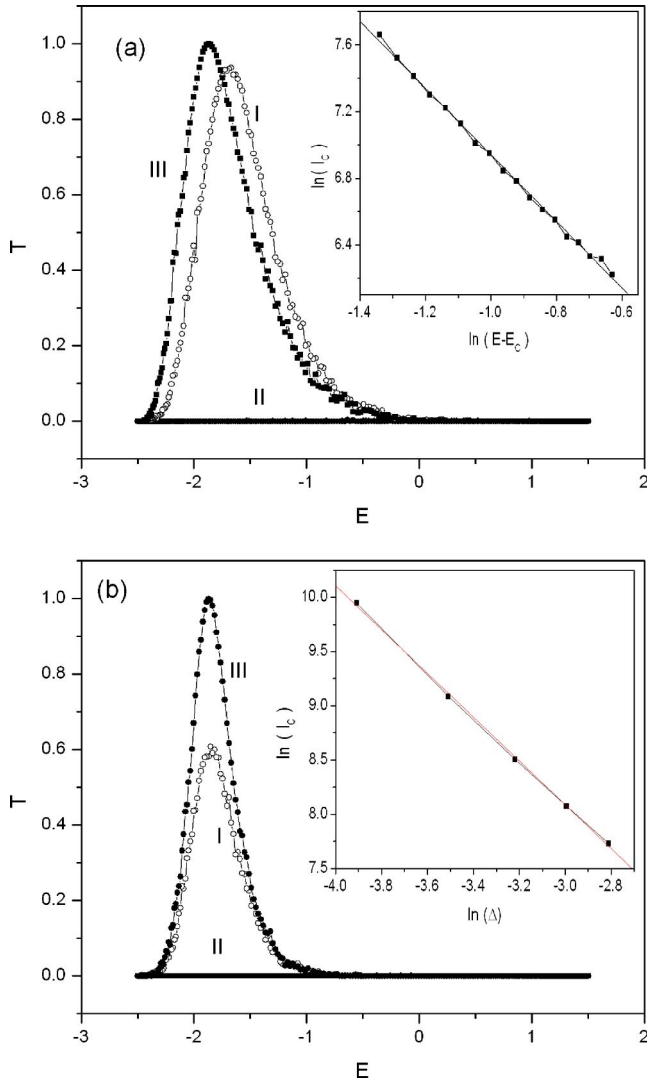
FIG. 1. (a) Transmission coefficients vs energy; $W=\varepsilon_B-\varepsilon_A$ $=1$. Curve I (empty symbols) is typical for system with local correlation; hopping constants here $t_{AA}=1$, $t_{BB}=1.73$, $t_{AB}=(t_{AA}+t_{BB})/2=1.36$. Curve II (solid line near zero) is for Anderson limit, with all hopping constants equal ($=1$). Curve III (solid symbols) is for system with "golden correlation," $t_{AB}=t_G\equiv\sqrt{t_{AA}t_{BB}}$ $=1.316$, with unit transmission at $E\simeq-1.9$. Concentration of ε_B site is 0.1 in all three curves. (b) Same as in (a), but with concentration of ε_B at 0.5. Inset in (a): Localization length $l(E)$ vs $(E-E_c)$ for system with golden correlation. Slope of fitted line is 2. Concentration of ε_B site is 0.5. Inset in (b): Localization length at critical energy $l_c=l(E_c)$ vs $D=(|t_{AB}-t_G|)$, where $t_G=\sqrt{t_{AA}t_{BB}}$ is the golden condition. Concentration of ε is 0.5.

figurations. For the purpose of comparison, we also show the transmission coefficient for the Anderson model (where $t_{AA}=t_{BB}=t_{AB}=t$) with the same degree of on-site disorder. We see that in *all* cases, the local correlation built-in through the t values leads to *much larger* transmission coefficient, compared with those in the Anderson model. When the concentration is small, there is in fact a regime of high transmission (~1), with energy centered around that given by Eq. (2).

With increasing B concentration, the electron has a higher chance to scatter from dimer and trimer impurities. For a single dimer impurity, a straightforward but cumbersome calculation yields the transmission

$$T_2(E)=\frac{(2t_{AA}t_{BB}t_{AB}^2\sin k)^2}{(2t_{AA}t_{BB}t_{AB}^2\sin k)^2+N_2^2},\qquad(3)$$

where $N_2=(t_{AA}^2-t_{AB}^2)(W-2t_{AA}\cos k)^2+t_{AB}^4-t_{AA}^2t_{BB}^2+t_{AB}^2W^2$ $-2Wt_{AA}t_{AB}^2\cos k$. In general, there are more energy values satisfying $T_2(E)=1$, but they are different from those satisfying $T_1(E)=1$, in general. Consequently, although short-range off-diagonal correlation leads to extended states for finite systems (localization length l_c larger than the system size L), these states are not extended states in the thermodynamic limit, $L\rightarrow\infty$.

Figure 1 also illustrates that for special local correlations there is an extended state even for infinite systems. One can easily verify that when $t_{AB}=t_G\equiv\sqrt{t_{AA}t_{BB}}$ (we call this the "golden correlation" in off-diagonal parameters), the condition $T_1(E)=T_2(E)=1$ can be satisfied. This implies that for the peculiar golden correlation t_G, single and dimer impurities are essentially transparent at this energy. The question remains how general is this result, that is, how about trimers or more general impurities? Instead of calculating $T_m(E)$ for m impurities, we prove the existence of an extended state by explicit construction. It is not difficult to check that under the condition $t_{AB}=t_G$, the state $\alpha_n e^{ikn}$, with $\alpha_n=1$ for $\varepsilon_n=\varepsilon_A$ and $\alpha_n=\sqrt{t_{AA}/t_{BB}}$ for $\varepsilon_n=\varepsilon_B$, is indeed an extended state with energy $E=E_c=\varepsilon_A+2t_{AA}\cos k=\varepsilon_B+2t_{BB}\cos k$. The physical picture for this state is then that the electron propagates on island A or B in the plane wave form, while the golden condition ensures perfect transition from island A to island B, and vice versa. One can say that this perfect transmission arises from the cancellation of backscattered waves produced by the subtle tuning of off-diagonal correlations. We find a state with unit transmission coefficient as that shown by curve III in Fig. 1(a), *even for high concentration of impurities*, although this "resonance" becomes sharper for high impurity concentrations [see Fig. 1(b)]. This is an example of an extended state in the thermodynamic limit in a random 1D system with short-range off-diagonal correlations (but *no* correlation in on-site energies). Notice also that $T(E_c)=1$ even for a system with 50% disorder, as shown in Fig. 1(b).

Under the golden correlation condition $t_{AB}=t_G$, the extended state satisfies $2\cos k=(\varepsilon_B-\varepsilon_A)/(t_{AA}-t_{BB})$, which can be met only when $|\varepsilon_B-\varepsilon_A|<2|t_{AA}-t_{BB}|$, resulting in an interesting effect. Usually in the presence of only diagonal or off-diagonal disorder, the larger the disorder is, the poorer is the transport. The situation is quite different for correlated diagonal and off-diagonal disorder. To obtain an extended state in the presence of the diagonal difference $W=\varepsilon_B-\varepsilon_A$, the difference between t_{AA} and t_{BB} has to be large enough, i.e., one needs the correlated off-diagonal disorder to be *large*. This is contrary to expectations.

Notice also that for fixed t_{AA}, and t_{BB}, there is a critical on-site difference $W=2|t_{AA}-t_{BB}|=2\Delta$. From the time evolution of a particle initially placed at a randomly chosen site (not show here), we find that when $W<2\Delta$, the mean-square displacement in time τ is $\langle x^2\rangle\sim\tau^{3/2}$, and then it is in a su-
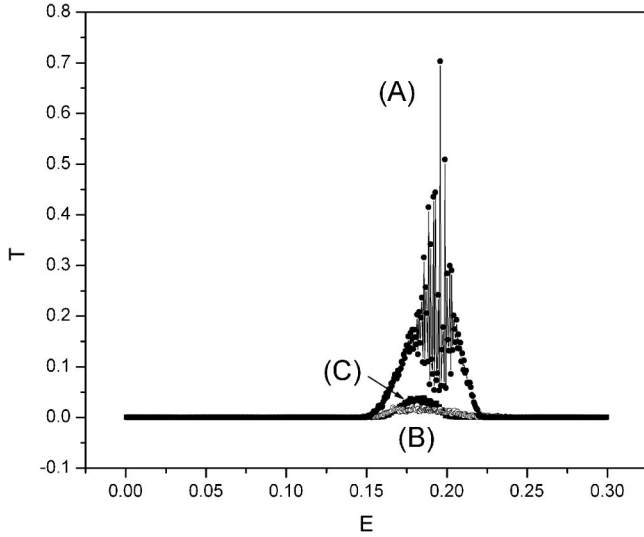
FIG. 2. Transmission for SL with 100 randomly distributed quantum wells of two types, width 2.6 (type a well) and 2.9 nm (type b). Barriers between same a (or b) wells have 3.6 nm (2.4 nm) width. Other barrier width is 3.0 nm (curve A, $\sim$ golden condition), 2.0 nm (curve B), and 3.8 nm curve (C). All barriers have height 0.3 eV. $T(E)$ averaged over 600 different disorder configurations. Concentration of b wells is 0.5.

perdiffusive phase. In contrast, when $W = 2\Delta$, the system is in a diffusive phase, $\langle x^2 \rangle \sim \tau$; and for $W > 2\Delta$, the mean-square displacement is bounded. This transition is similar to that in the random dimer model (RDM), although with different characteristics.[2] In RDM, the transition occurs at $W = 2t_{AA}$ (all t the same). In our case, the condition is related to the difference between hopping constants, and not the hopping constants themselves. It is interesting that for $t_{AA} = t_{BB} < W/2$, there are extended states in RDM, but no extended state in our model.

We study the localization length $l(E)$ for states near the critical energy E_c in Fig. 1(a) inset. We find that $l(E) \propto (E - E_c)^{-2}$ for states near E_c. The number of extended states for a system of length L [i.e., $l(E) > L$] is related to $\delta k \propto E - E_c \propto L^{-1/2}$, where near E_c, $E = E_c + A\,\delta k$. The number of extended states is then $\delta k / (1/L) = L^{1/2}$, a sizable number, just as in the RDM.[12]

The long-time behavior of the system is determined by a critical exponent. One can show the relation between two exponents θ and γ, defined by $\langle x^2 \rangle \sim \tau^{\theta}$ and $l(E) \sim |E - E_c|^{-\gamma}$. For short times, the electron has ballistic behavior, since it has not sampled yet the disorder potential, so that $\langle x^2 \rangle \sim (v\tau)^2$. For long time, however, $\langle x^2 \rangle \sim l^2(E)$, for an electron with energy E. We can then write the mean-square displacement as $\langle x^2 \rangle = \int dE \rho(E)(v\tau)^2 f(v\tau/l(E))$, where $\rho(E)$ is the density of states, and we surmise the scaling function $f(x) \to 1$, as $x \to 0$, and $f(x) \to 1/x^2$, as $x \to \infty$. From this, one obtains $\langle x^2 \rangle \sim \tau^{2-1/\gamma}$, for long times, so that $\theta = 2 - 1/\gamma$. When $\gamma = 2$, as in Fig. 1(a) (inset), $\theta = \frac{3}{2}$ (superdiffusive regime); while when $\gamma = 1, \theta = 1$ (diffusive). There is perfect agreement with our numerical calculations.

It is natural to expect that in many systems there exist correlations between diagonal and off-diagonal disordered

parameters. However the golden correlation condition is not necessarily satisfied, and it is important to see how the transport properties change when a system deviates from this. The inset in Fig. 1(b) shows that $l(E_c) \propto (t_{AB} - t_G)^{-2}$, so that to obtain extended states, we need $|t_{AB} - t_G| < L^{-1/2}$. As long as this condition is met, effective conduction channels are opened by the off-diagonal correlations in the disordered system.

Our predictions could be verified experimentally in systems with access to varying degree of disorder and structural correlation, such as model semiconductor superlattices (SL's).[13] Consider a SL with quantum wells of two different widths d_A and d_B, distributed randomly in the structure. The barriers between wells have the same height U (given by the material composition) and width b_A (or b_B) if the barrier is between two alike wells of width d_A (or d_B), and otherwise have widths b_C. An estimate of the hopping constant between two quantum wells with width d_L and d_R, separated by a barrier of width b and height U, is $t = (\pi^2 \hbar^2 / ms)\sqrt{d_L d_R}\exp(-sb)$, where $s = \sqrt{2mU}/\hbar$. By tuning parameters, the golden condition can be attained. Figure 2 shows the transmission for different systems calculated from a Kronig-Penney model of the SL. Curve A is for a system satisfying the golden condition, as estimated from the expression above, while curves B and C are results away from the condition. The discussion above for the tight-binding model suggests that transport would indeed be better for system in curve A, even as the barrier between different quantum wells (curve B) is *thinner*. We emphasize that Fig. 2 is obtained from a Kronig-Penney model of the structure, so that hopping amplitudes go far beyond nearest neighbors, and the golden condition is likely much more involved than in the tight-binding model. The golden condition for curve A was *not* optimized, but just estimated from the relation above, and the difference between these curves is remarkable.[14]

As discussed before, our studies have direct application to models of transport in DNA in the literature.[15,16] For a typi-
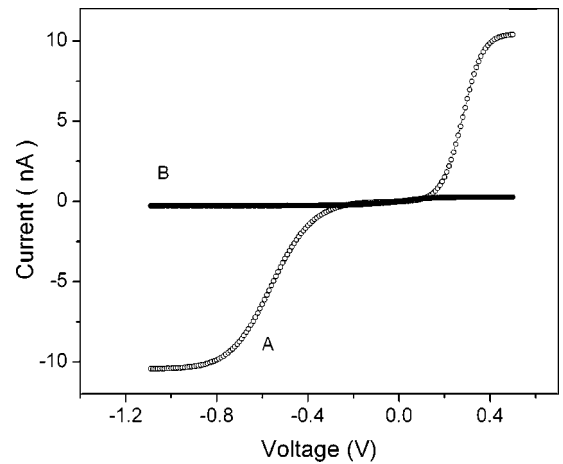

FIG. 3. *I-V* curves for a random base pair sequence (i.e., random on-site energies). Curve A is for model of λ-DNA with realistic local correlation in the hopping amplitudes. Curve B is for random diagonal Anderson model with hopping amplitudes set equal. Size of systems is 562; temperature in Fermi broadening is 300 K.

cal DNA molecule the base pair sequence may be essentially random, such as in λ-DNA. However, the chemical structure determines the local correlation between on-site energies and hopping constant via the π-orbital overlap. In order to explore how the local correlation changes transport, we compare the I-V curves of different systems, obtained using the Landauer-Büttiker formalism,[17] $I=(2e/h)\int dE\,T(E)[f_L(E)-f_R(E)]$, where $f_{L/R}(E)=\{\exp[E-\mu_{L/R}/k_BT]+1\}^{-1}$ is the Fermi function. We choose $\mu_L=E_F+(1-\kappa)eV$ and $\mu_R=E_F-\kappa eV$, where E_F is the equilibrium Fermi energy, V is the applied voltage, and κ is a parameter describing the possible asymmetry of contact to leads, chosen here as $\kappa=1/3$.[18,19] We assume that the DNA is attached to ideal leads described by a metal with bandwidth 1.2 eV. The hopping constant between leads and DNA chain is chosen to be $\sim t_{AB}/10\simeq 0.01$ eV, reflecting a relatively poor contact. We use two different sets of parameters: $t_{AA}=-0.0695$ eV, $t_{BB}=-0.1409$ eV, and $t_{AB}=(t_{AA}+t_{BB})/2$, curve A in Fig. 3, describe a realistic molecule, as the values are obtained from microscopic calculations;[10] $t_{AA}=t_{BB}=t_{AB}=-0.1403$ eV, curve B, simulates an uncorrelated system, i.e., the Anderson limit.

We can see from Fig. 3 that the current in the system with local correlation (curve A) is overall much larger than in the system without correlation, even though the hopping constant is larger in B. There is in fact no conductance over the entire bias range for curve B, with no correlation in the hopping constants. The message of these results is that even for DNA with random sequences, such as λ-DNA, "good" transport is possible due to the effective conduction channels opened by structural correlations. Notice that t_{AB} in curve A does not satisfy the golden condition (~-0.099 eV) by about 4%, and yet, there is significant current amplitude for finite biases. In contrast to the conducting states in polymers, which arise from the correlation in local energies, the conducting states here have to do with correlation in hopping amplitudes. It is clear that the backbone may change the local correlations. We may conclude that changes in local correlation will lead to changes in the I-V features, which may in fact be an ingredient in recent experiments, especially if chemical changes affect the molecule structure.[20]

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[1] E. Abrahams, P.W. Anderson, D.C. Licciardello, and T.V. Ramakrishnan, Phys. Rev. Lett. **42**, 673 (1979).

[2] D. Dunlap, H.-L. Wu, and P. Phillips, Phys. Rev. Lett. **65**, 88 (1990); P. Phillips and H.-L. Wu, Science **252**, 1805 (1991).

[3] F.M. Izrailev and A.A. Krokhin, Phys. Rev. Lett. **82**, 4062 (1999); F.M. Izrailev, A.A. Krokhin, and S.E. Ulloa, Phys. Rev. B **63**, 041102 (2001).

[4] C. Timm, F. Schafer, and F. von Oppen, Phys. Rev. Lett. **89**, 137201 (2002).

[5] N. Sandler, H.R. Maei, and J. Kondev, Phys. Rev. B **68**, 205315 (2003).

[6] T. Heinzel *et al.*, Europhys. Lett. **61**, 674 (2003).

[7] M. Di Ventra and M. Zwolak, *"DNA Electronics," Encyclopedia of Nanoscience and Nanotechnology*, edited by H.S. Nalwa (American Scientific, Stevenson Ranch, 2002).

[8] E.M. Conwell and S.V. Rakhmanova, PNAS **97**, 4556 (2000).

[9] P. Carpena, P. Bernaola-Galvan, P.Ch. Ivanov, and H.E. Stanley, Nature (London) **418**, 955 (2002).

[10] R.G. Endres, D.L. Cox, and R.R.P. Singh, cond-mat/0201404 (unpublished).

[11] π-orbital chains for the corresponding two base pairs per "rung," as well as the poorly-conducting phosphate backbone chain, can all be reduced into an effective 1D model with renormalized on-site energy and hopping constant (Ref. 15). The effective constants are those obtained from first-principles calculations which estimate bandwidths in these systems (Ref. 10).

[12] Note that near the band edge, there is an anomaly, and $(\delta k)^2 \propto E-E_c \propto L^{-1}$, but the number of extended states is still $\propto L^{1/2}$.

[13] V. Bellani *et al.*, Phys. Rev. Lett. **82**, 2159 (1999).

[14] Despite averaging over 600 configurations, curve A in Fig. 2 exhibits large oscillations for $E\simeq E_c$, reflecting the sensitivity of these states to boundaries and disorder, which produces poor self-averaging and arises from their extended nature.

[15] G. Cuniberti, L. Craco, D. Porath, and C. Dekker, Phys. Rev. B **65**, 241314 (2002).

[16] Z.G. Yu and Xueyu Song, Phys. Rev. Lett. **86**, 6018 (2001).

[17] R. Landauer, Philos. Mag. **21**, 863 (1970).

[18] X.Q. Li and Y.J. Yan, Appl. Phys. Lett. **79**, 2190 (2001).

[19] S. Datta, W. Tian, S. Hong, R. Reifenberger, J.I. Henderson, and C.P. Kubiak, Phys. Rev. Lett. **79**, 2530 (1997).

[20] B. Hartzell *et al.*, Appl. Phys. Lett. **82**, 4800 (2003); J. Appl. Phys. **92**, 2764 (2003).