

Ballistic to diffusive transition in a two-dimensional quantum dot lattice

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Two-dimensional networks of ordered quantum dots beyond the percolation threshold are studied, as a typical example of conducting nanostructures with quenched random disorder. Theory predicts anomalous diffusion with stretched-exponential relaxation at short distances, and computer simulations on lattices of crossing, straight paths of random length confirm such a behavior. Anomalous diffusion is interpreted as resulting from the higher probability of taking straight, or ballistic, paths, when the traveled distance is comparable or shorter than the lattice characteristic length. Diffusion turns over to normal for longer traveled distances, whence all paths tend to become equiprobable. Such random lattice structures may represent a model for realistic quantum dot networks, with potential applications in optoelectronics, photovoltaics, or spintronics.

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I. INTRODUCTION

Network-like structures made from one-dimensional (1D) nanowires, nanorods, or nanotubes as building blocks can function both as devices and interconnects, and are thus expected to play a prominent role in next-generation nanotechnology. Recently, the synthesis of several such structures in two or three dimensions (2D, 3D) by different techniques was reported for, e.g., CdS, WO₃, InAs, and PbSe nanowires [1–3]. Applications can range from electronics [4] to electrochemistry [5] to strain monitoring [6], and so forth. However, very intriguing 2D networks can also be synthesized by the self-assembly of arrays of quantum dots into superstructures, thus obtaining networks that can range from perfectly ordered to quite disordered, over different length scales [7–10]. Such quantum dot superlattices display peculiar electronic band structures [11], effectively behaving as arrays of pseudoatoms with discrete states combined into bands. Electron injection, tunneling, and hopping through these nanostructures becomes possible, and such “metamaterials” are predicted to have a strong potential for optoelectronic, photovoltaic, and spintronic applications.

In this work, I study a general problem of determining the conductivity of carriers (“electrons”) hopping between sites occupied by a disordered array of conducting reservoirs (“quantum dots”), distributed over a planar region. The treatment is semiclassical, by defining a temperature and density range for which quantum correlation and conductance quantization effects can be ignored. From a more fundamental viewpoint, such structures are also good candidates for studying anomalous diffusion, because of the mixing of transport pathways with largely different probabilities. Non-Fickian diffusion and stretched-exponential correlation functions often arise as a characteristic feature of transport in strongly inhomogeneous media, in such diverse systems ranging from cell membranes to groundwater flow [12–15]. The underlying physics may be reduced to the random walk over a two-dimensional, multiply connected lattice containing traps at some sites, although in our case (see below) the role of “traps” must not be taken literally. In the following, I will

first derive an analytic approximation for the probability of traveling a path of given length, based on the diffusion over a set of broken domains. Then, a computer model is formulated for the random walk on a superpercolating 2D lattice, by constructing random networks of crossing segments with variable aspect ratio. Computer simulations of the traveled path length and traveled straight distance allow one to deduce the asymptotic behavior of the hopping current in such disordered networks, supporting the theoretical prediction of stretched-exponential anomalous diffusion over distances shorter or comparable to the lattice characteristic correlation length, indicating ballistic transport, while diffusion turns over to normal at larger distances.

II. THEORETICAL MODEL

Let us start from a simple $N \times N$ square lattice with z -fold connectivity. Each site, described by a position \mathbf{r} and nearest-neighbor distance $d = \mathbf{r} - \mathbf{r}'$, can be occupied by a quantum reservoir, which may be charged by a variable number of carriers. To fix the ideas, the reservoir could be identified with a semiconductor quantum dot and the carriers with electrons. The carrier density is taken to be not high, such that correlated diffusion of several electrons through adjacent paths is negligible; i.e., we consider a single-particle diffusion picture in which quantum correlation effects can be ignored. The energy scale for charging a dot with a single electron can be simply estimated as $E_c = \frac{e^2}{4\pi\epsilon\epsilon_0 d} \sim \frac{1.44}{\epsilon d}$ eV, with d in nm, and ϵ typical values ranging from 4, e.g., in CdSe quantum dot films [16], to ~ 12 – 13 in silicon or GaAs heterostructures.

The site occupation rate of such a lattice must be beyond the percolation threshold to permit long-range diffusion of the carriers. However, the presence of geometrical correlations between the sites (i.e., crossing straight paths of variable length $Q < N$) makes the percolation threshold depend on the “aspect ratio,” i.e., the average length of straight paths for a given occupation density. Note that, besides dots hopping, this setup is also representative of the transport across a 2D network of randomly dispersed conducting nanowires or nanotubes, with average sizes smaller than the characteristic length ($\sim \sqrt{A}$ for an occupied surface area A).

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At very low temperatures, the process of single-electron hopping was predicted by Mott to go beyond the simplistic nearest-neighbor jump between localized sites: diffusing electrons rather look for the best compromise between the lowest activation energy and the shortest jump distance. Mott’s variable-range hopping gives an exponential relationship between conductance and temperature, $G \propto \exp(-B/T^{1/3})$ in two dimensions (in 3D the temperature exponent changes to 1/4). At higher temperatures, however, the jump activation energy becomes significantly smaller than the dot charging energy, and the network conductance starts following an Arrhenius-like diffusive behavior [17,18], thereby signaling the prevalence of simple nearest-neighbor hopping (i.e., finding the shortest path becomes the rule). For a generic assembly of dense quantum dots, the transition temperature can be estimated as

$$T_0 = \left(\frac{a}{4d}\right)^2 T^* \quad (1)$$

and $T^* \simeq 2.8(E_c/k_B)(d/a)$. The quantity a is the localization length, which is typically a factor of 2–5 smaller than the dot size d . For d in the range of a few nm, the charging energy is a few tens of meV, and the transition temperature is $T_0 \sim 10\text{--}100$ K, depending on the material and dot size d (the smaller the dot, the higher the T_0). Therefore, at about room temperature nearest-neighbor hopping dominates the physics of carrier transport, and single-particle random hopping across the filled lattice sites will be the only mechanism considered in this work. Also note that at low temperatures, quantum effects such as Coulomb blockade can be observed, while at room temperature electrons thermally overcome the Coulomb barrier between dots (except for extremely small dots, for which discrete conductance may still be observed up to near room temperature [19]).

As said, the density of electrons is assumed to be small, so that also direct e-e collision or dot saturation can be neglected. Therefore, one may focus on the random walk of a single particle. As usual in this kind of problem [20,21], we look for the probability $\overline{W}(\mathbf{r},t)$ that after t steps the walker is found at the lattice position \mathbf{r} . The equation of motion for the probability is

$$\overline{W}(\mathbf{r},t+1) = \sum_{\mathbf{r}'} p(\mathbf{r}-\mathbf{r}') (1-\delta_{\mathbf{r}'}) \overline{W}(\mathbf{r}',t) + \delta_{\mathbf{r}} \overline{W}(\mathbf{r},t). \quad (2)$$

$p(\mathbf{r}-\mathbf{r}')$ is the (geometric) probability of transition from a site \mathbf{r}' to a neighboring site \mathbf{r} in the dense lattice (without traps, or dead ends); for a fixed lattice geometry and connectivity it is just a constant, equal to some effective “diffusion coefficient” D , the p are inversely proportional to the lattice coordination z , and the sum of all the p must equal unity. The switch function $\delta_{\mathbf{r}}$ is 0, except at those sites \mathbf{r} representing a trap, where it is equal to 1 (therefore arresting the random walk); in practice, it is the matrix of lattice sites occupied by a dot (=0) or empty (=1). The last term in the equation is the (final) contribution of a trapped particle to the equation of motion. From this writing, it is seen that the probability $\overline{W}(\mathbf{r},t)$ is indeed independent of the time (conservation of the norm). By assuming a finite mobility, the time variable can be made to correspond with a traveled path length $Q = vt$ at constant velocity v .

As noted above, the meaning of “traps” for our lattice is not just that of a zero-mobility site where, in the absence of recombination, charge would accumulate. In this picture, an electron starts from a dot, and keeps hopping randomly to neighboring dots, until it attempts jumping to an empty site, representing a dead end for the walking electron. In disordered systems momentum is conserved only on average, and it can be considered that at each hop the electron linear momentum is randomized. In fact, the hopping carrier behaves as a Brownian particle, which chooses at each step a new value of momentum. However, when reaching a “trap,” its choice becomes obligatory; it can only reflect \mathbf{k} into $-\mathbf{k}$ thus inducing an unwanted correlation in the statistics. From the practical point of view of computing statistical path-length distributions of various kinds (to be defined later on), it can be considered as a “new” electron: its trajectory is interrupted and restarted at another random site. Only in this sense, dead ends represent a trap.

In fact, we are interested only in knowing the values of the probability $\overline{W}(\mathbf{r},t)$ for the sites with $\delta_{\mathbf{r}} = 0$, i.e., for the probability of a walker *not* falling into a trap. Then, by summing up over all the paths not ending in a trap, an estimate can be obtained of the probability for an electron to travel over a distance, as a function of the lattice connectivity. Therefore, it is useful to introduce the auxiliary probability:

$$W(\mathbf{r},t) = (1-\delta_{\mathbf{r}}) \overline{W}(\mathbf{r},t). \quad (3)$$

This coincides with \overline{W} at the filled sites, and vanishes at trap (empty) sites. By following [20,21], its equation of motion is obtained by multiplying the (1) by $1-\delta_{\mathbf{r}}$:

$$W(\mathbf{r},t+1) = D \sum_{\rho} \eta(\mathbf{r},\mathbf{r}+\rho) W(\mathbf{r}+\rho,t), \quad (4)$$

where $\eta(\mathbf{r},\mathbf{r}+\rho) = 1$, if both \mathbf{r} and ρ are occupied, and 0 otherwise. It is a standard procedure to obtain the formal solution to the (3) above by setting it as an eigenvalue problem:

$$D \sum_{\mathbf{r}'} \eta(\mathbf{r},\mathbf{r}') W(\mathbf{r}+\rho,t) \phi_n(\mathbf{r}') = \lambda_n \phi_n(\mathbf{r}). \quad (5)$$

As shown in [21], the general solution is expressed in terms of the initial distribution $W_0(\mathbf{r})$ as

$$W(\mathbf{r},t) = \sum_n A_n \phi_n(\mathbf{r}) \lambda_n^t, \quad (6)$$

$$A_n = \sum_{\mathbf{r}} \phi_n^*(\mathbf{r}) W_0(\mathbf{r}). \quad (7)$$

Note that at $t = 0$ the (6) becomes an identity because of the orthonormality of the ϕ_n . Balagurov and Vaks presented a general solution for this problem by a spectral method, first introduced by Lifshitz [22].

However, an easier way to understand the behavior of the solutions is to first note that Eq. (4) in *one* dimension reduces to the ordinary diffusion equation over a piecewise connected domain of total length L . The traps are represented by i randomly distributed empty sites, corresponding to a concentration c . Each trap i delimits a portion on the segment of length $l_i = |x_i - x_{i-1}|$, with x_0 the origin and $x_{c+1} = L$. The lengths l_i can be arbitrarily distributed to reflect the presence (or absence) of spatial correlations in the lattice (1D domain

in this case). The probability is subject to $W(x_i, t) = 0$ for $i = 0, \dots, c + 1$ at any t , and $W(0 < x < L, t = 0) = 1/L$. In this case, the general solution would be the well-known [23]

$$W(x_i, t) = (4/L) \sum_n \exp(-k_n^2 Dt/2) \frac{\sin k_n(x - x_i)}{k_n l_i} \quad (8)$$

for $k_n = (2n + 1)\pi/l_i$.

The probability of traveling freely over a time t (or a total path length $Q = vt$) is the average of integrals of $W(x, t)$ over the segment lengths delimited by the random distribution of traps:

$$P(t) = \sum_i \left\langle \int_{x_i}^{x_{i+1}} W(x, t) dx \right\rangle, \quad (9)$$

the $\langle \dots \rangle$ indicating averaging of all possible distributions of the segment lengths l_i .

The term k_0 in the sum (8) defines the smallest wave number of the walker, whose inverse length defines the largest size, surface area, or volume (in 1D, 2D, or 3D) within which the walker will *not* be captured by a trap. One can exploit the analogy between the 1D solution of the diffusion equation on a piecewise continuous domain (8), and its 2D (or 3D) analogs, by replacing the limiting segment by a limiting circle (or sphere).

By restricting the discussion to $k_0 = \pi/l$, and taking a Poisson distribution of traps, $p(S) = c \exp(-cS)$, with concentration c , over a circle of surface $S = 4/k_0^2 = 4\pi l^2$, the average of W is given by

$$P(t) = \frac{\int_0^\infty p(S)W(S, t) dS}{\int_0^\infty p(S) dS} \propto \int_0^\infty c e^{-cS} e^{-\pi^2 Dt/S} dS, \quad (10)$$

that is, an integral of the type

$$I(t) = \int_0^\infty c e^{-cS - \pi^2 Dt/S} dS \\ = 2\pi(Dct)^{1/2} K_1(2\pi\sqrt{Dct}), \quad (11)$$

with K_1 the modified Bessel function. At long times, $K_1 \sim \exp(-2\pi\sqrt{Dct})/(Dct)^{1/4}$; therefore $P(t)$ decreases as a stretched exponential:

$$P(t) \sim (Dct)^{1/4} e^{-2\pi(Dct)^{1/2}}. \quad (12)$$

On the other hand, for a flat distribution $p(S) = 1/c$ (a “gas-like” distribution of traps) the long-times solution goes rather as a standard (diffusion-like) exponential:

$$P^*(t) \sim e^{-\pi^2 Dct}. \quad (13)$$

The above results for the probability of free-travel time qualitatively coincide, apart from numerical factors of order 1, with the results of Balagurov-Vaks [21] and Ryazanov [20]. Notably, sublinear diffusion near the percolation threshold has been often invoked to characterize the random walk over a complex (heterogeneous) configuration space [24–26].

III. NUMERICAL SIMULATIONS

To verify the above asymptotic limits, I set up a simulation model by filling up a square $N \times N$ lattice with straight segments made up of rows of “dots.” The lattice has a typical

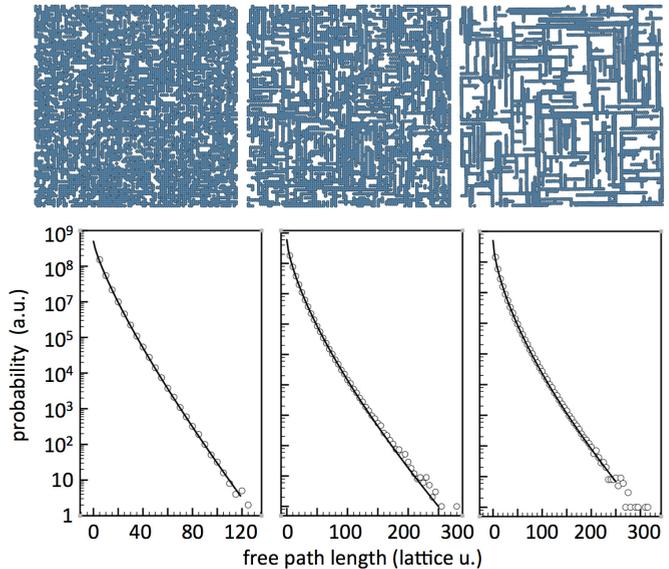


FIG. 1. (Color online) Top row: Schematic representation of a portion of two-dimensional 500×500 square lattices, filled with straight segments of dots, with aspect ratio $\bar{a} = 6$ (left, in lattice units), 12 (middle), and 18 (right); the aspect ratio is defined by the average number of adjacent dots in a random segment. Bottom row: Plot of the path-length probability $P(Q)$ for the three values of the segment aspect ratio $\bar{a} = 6$ (left), 12 (center), 18 (right). Continuous lines represent fits with stretched-exponential law, with exponents $\alpha = 0.8, 0.7, 0.65$, respectively.

size of $N = 500$. Using an exponential filling probability, straight segments of dots along the x and y direction can be built, with variable segment length a . For a short average segment length \bar{a} (or “aspect ratio”) the segment-length distribution is flat, while it becomes increasingly closer to a Poissonian as \bar{a} approaches the lattice length size. Therefore, by continuously varying the aspect ratio of the segments, the range from a flat to a fully Poisson distribution can be explored. Three examples of lattices built with this procedure are shown in Fig. 1 (top row): such configurations could be taken as an idealization of real experimental structures; see, e.g., Fig. 1 of Ref. [9].

A random walker starts from an occupied site and proceeds by jumping to neighbor occupied sites, until a move brings it to an empty site (a “trap”), at which point the walk stops, and a new walker is launched. Typical simulation runs are realized with $m \sim 10^6 - 10^7$ walkers. Periodic boundaries are applied throughout. In this way, statistics about the free-travel time, and therefore the traveled free-path length Q , can be accumulated, by averaging the contributions $p_i(Q), i = 1, \dots, m$, for each given occupation density and segment aspect ratio. In Fig. 1 (bottom row), the results for the distribution $P(Q) = \frac{1}{m} \sum_{i=1}^m p_i(Q)$ [hereafter indicated as $\langle p_i(Q) \rangle$ for brevity] are shown for different aspect ratios $\bar{a} = 6, 12, 18$ (in units of the lattice mesh), together with stretched-exponential fits of the type $P(Q) = A \exp(-BQ^\alpha)$. The best fit for the stretching exponent gives $\alpha = 0.8, \alpha = 0.7, \alpha = 0.65$, for the three aspect ratios, respectively. Hence, it appears that geometrical correlations in the segment length allow one to numerically span the range of probability distributions

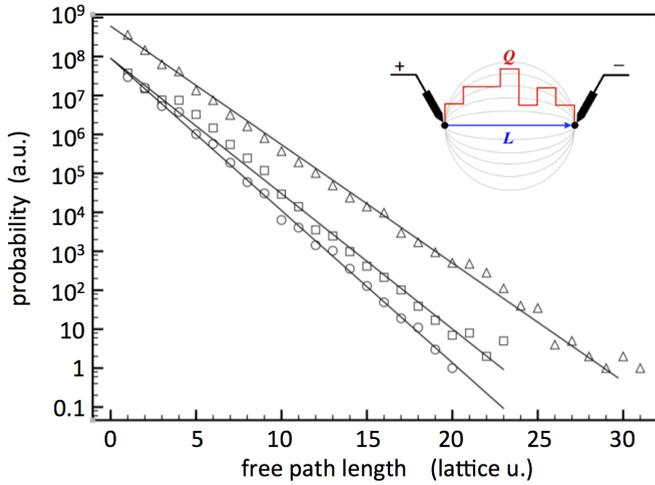


FIG. 2. (Color online) Plot of the traveled distance probability $P(L)$ for the three values of the segment aspect ratio $\bar{a} = 6$ (circles), 12 (squares), 18 (triangles). Continuous lines represent exponential law fits, with coefficients $\beta = 0.9, 0.8, 0.7$, respectively. The inset shows the difference between the path length Q (red) and the traveled distance L (blue), which may be measured under the application of a potential at two points in the lattice. The shape of the electric field lines is depicted in gray.

analytically determined by the extremes $P^*(t)$ and $P(t)$, respectively approaching an exponent of 1 as in Eq. (13) for the shorter aspect ratio (in the limit of $a \rightarrow 1$, the gas-like random distribution is recovered), and an exponent of 0.5 as in Eq. (12) for an increasing aspect ratio.

If now, rather than on the total traveled path length, we focus on the traveled *distance*, that is, the straight distance L between the end points of each free path, the plots shown in Fig. 2 are obtained. This probability distribution is calculated by adding all the contributions from any path leading to a same value of L , for a random distribution of starting points. The distribution $P(L) = \langle p_i(L) \rangle$ is clearly exponential, $P(L) \sim \exp(-\beta L)$ with a coefficient proportional to the effective diffusion coefficient, $\beta = 0.9, 0.8, 0.7$, respectively, for the aspect ratios $\bar{a} = 6, 12, 18$.

Note that the traveled distance is the important quantity when looking at the particle current. For example, in the case of electrons jumping through charged dots, the driving force to push the electrons from one point in the lattice to another one at a distance L would be provided by an electric field. Experiments of such kind may be performed by placing nano electrodes at two contact positions separated by L (see inset in Fig. 2), and shooting a voltage difference between the two tips [27]. For a fixed polarization voltage V , the electric field scales as $E \propto L^{-1}$. Under such a condition, electrons will flow by taking all the possible paths of length Q , leading from 0 to L , and the current will result from a weighted average over all such paths.

A probability function for electrons traveling the different paths leading to the same distance under a driving force can be constructed by observing that the electric field between two points at distance L , and at different potential, roughly decreases as L/Q (see again inset to Fig. 2). This probability $P_L(Q)$, proportional to the electron current, can be calculated

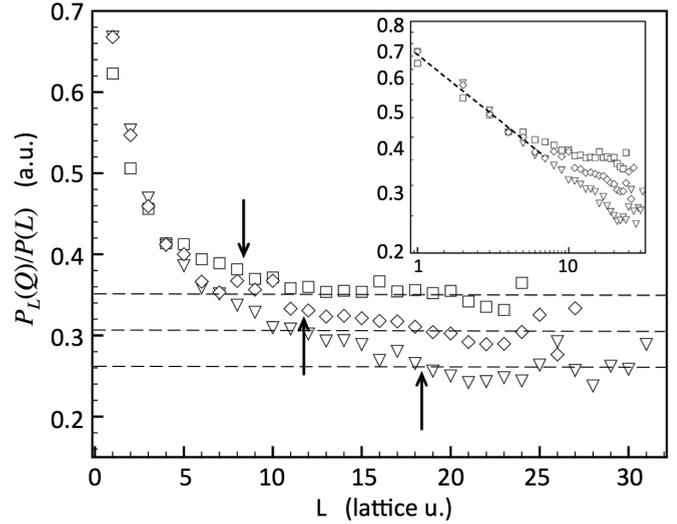


FIG. 3. Plot of the ratio of field-weighted probability to traveled distance probability $P_w(L)/P(L)$ for the values of the segment aspect ratio $\bar{a} = 6$ (squares), 12 (diamonds), and 18 (triangles). Dashed lines represent the respective asymptotic constant values at large L . Arrows indicate the approximate value of L_{sat} at which saturation to a constant value occurs. The inset shows the same data on a log-log scale.

by weighting each contribution $p_i^L(Q)$ to the statistical distribution $P(Q)$ by the factor L/Q . [The superscript L indicates that only the paths Q ending at the same straight distance L are counted in $P_L(Q)$.] To normalize the result to one electron, i.e., to a current density j , I plot in Fig. 3 the ratio $P_L(Q)/P(L)$, for the different values of aspect ratio $\bar{a} = 6, 12, 18$. It is easily seen that this ratio must be proportional to the effective carrier mobility, since

$$\frac{P_L(Q)}{P(L)} = \frac{L \langle \frac{1}{Q_i} \times p_i^L(Q) \rangle}{\langle p_i(L) \rangle} \propto \frac{j}{E}. \quad (14)$$

For values of L large compared to a , the effective mobility per electron saturates to a constant value, whereas at shorter distances (i.e., a closer distance between the tips of the nano electrodes) it increases faster than linearly. The finite lattice size does not allow one to estimate the divergence for infinitely close tips, where the electric field would approach infinity; however, in real experiments the dots obviously have a finite size, which here is represented by the lattice unit mesh.

IV. DISCUSSION AND CONCLUSIONS

At short distances, most hopping paths are straight or nearly straight, i.e., ballistic rather than diffusive. At large distances, $L \gg \bar{a}$, the current (flow probability per electron) becomes constant as it should, in a macroscopic condition. In the framework of our disordered quantum-dot network it is understood that, once L is larger than the average segment length, no straight paths (ballistic) from 0 to L are possible, while at increasing L all the long electron tunneling paths with $Q \gg L$ (diffusive) tend to become equiprobable. It can be also observed (see the arrows in Fig. 3) that the value of L_{sat} at which the saturation to a constant value occurs corresponds

quite well to the aspect ratio of the segments filling the lattice (i.e., the average length of the straight segments), $L_{\text{sat}} \simeq \bar{a}$.

In the hopping regime, the Nernst-Einstein equation linearly relates the conductivity (i.e., the mobility) to the diffusion coefficient, $\sigma = e^2 D / k_B T$. From Fig. 3, it can be seen that the asymptotic saturation value of the P_L/P ratio actually scales with the inverse of the network aspect ratio; i.e., the effective mobility in the diffusive regime goes as $\mu \propto 1/\bar{a}$. This is indeed consistent with the previous observation, according to which the coefficient β in the exponential distribution of $P(L)$, proportional to the carrier diffusion coefficient D , was found to decrease linearly upon increasing \bar{a} .

However, as shown in the inset of Fig. 3 where the same data for P_L/P are plotted on a log-log scale, in the path-length domain $L \lesssim \bar{a}$, the mobility turns out to be independent of the lattice characteristic length \bar{a} , and rather takes on a power-law dependence on the path length, $\mu \propto L^{-\xi}$, with a weak exponent $\xi \simeq 0.1$. Since this transition coincides exactly with the change of behavior from diffusive to ballistic, as well as with setting in of the anomalous diffusion with stretched-exponential decay (see Fig. 1), one should take such a peculiar power-law behavior of the mobility as characteristic of the ballistic regime. Indeed, it may still be appropriate to retain the concept of mobility also in this regime, since in the absence of a driving force no net transport would occur. However, the rather small value of the exponent ξ likely suggests that this regime could be rather a sub-ballistic one [28], for which the rms diffusion length is intermediate between the purely diffusive, $\langle \mathbf{r}^2 \rangle \propto t$, and truly ballistic, or Taylor, regime [29], whence $\langle \mathbf{r}^2 \rangle \propto t^2$.

In conclusion, in this work I developed a semiclassical model of carrier hopping through a disordered lattice of charge reservoirs, or quantum dots, in a temperature and density regime for which purely quantum effects, such as correlated multiparticle transport or quantized conductance, can be ignored. A simplified theoretical formalism shows that by mapping this problem over that of classical particles

diffusing over a lattice of connected segments of finite length, stretched-exponential scaling can be observed for the path-length distribution function. Transition to ordinarily diffusive hopping transport is recovered when the path length exceeds the correlation length of the lattice. Numerical simulations confirm these predictions, and allow one to deduce some interesting while more general conclusions.

First, the stretched-exponential behavior appears not merely as a convenient fitting function, but naturally arises from the distribution of free-path segment lengths. Whenever straight, or nearly straight, paths connecting two points in a disordered network of conductors are available to carriers, these will travel ballistically the distance in a time shorter than the average classic (Fickian) diffusion time. This leads also to a slower decay of the distance autocorrelation function, a phenomenon often observed near the percolation threshold for various physical systems.

Second, the current measured between two random sites in a disordered network of conductors can increase faster than linearly, when the average length of the conducting elements (such as nanowires, nanotubes, arrays of conducting dots) is comparable to or larger than the distance between the two sites. In other words, the conductance between the two points becomes a nonlinear function of the distance, because of the relative dominance of ballistic over diffusive pathways at short distances.

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