Transport of Electron-Hole Pairs in Arrays of Small Tunnel Junctions

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We investigate charge transport in two capacitively coupled 1D arrays of small tunnel junctions, attached to two different circuits. The electron and hole placed in adjacent electrodes of different arrays form a sort of "exciton" that moves as a whole. It was shown that under certain conditions only these excitons contribute to the charge transport along the junction arrays, so that the currents in the circuits are exactly equal in magnitude and opposite in direction. The existence of these excitons provides an interesting possibility to simulate some features of microscopic excitons in a mesoscopic system.

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Recently, the systems of submicron metal tunnel junctions became the subject of considerable interest due to the Coulomb-blockade effects that provide the possibility to manipulate single electrons.

The physical background of these effects is the simple semiclassical picture of electron transport called the "orthodox" theory. It is valid if the junction resistances are high enough: \( R_T \gg R_Q = \frac{\pi \hbar}{2e^2} \approx 6.5 \, \text{k} \Omega \). In the framework of this theory there are well-defined numbers of electrons, i.e., charges, on the junction electrodes. These charges are changed in the process of single-electron tunneling. Such a tunneling can occur at zero temperature only if it is energetically favorable with respect to the electrostatic energy of the system. If the resistance of the junction electrodes are much smaller than \( R_Q \) (which is typically the case), it is the electrostatic energy of the whole system that is relevant for tunneling.

When the single-electron processes can only increase the electrostatic energy and, hence, are forbidden, more complex elementary processes that involve several electron-tunneling events become dominant. These processes are essentially quantum mechanical and go through virtual intermediate states. Hence, the tunneling of one electron cannot be separated from the tunneling of another electron involved in such a multielectron process. For this reason we prefer to call these processes cotunneling to distinguish them from the processes of correlated tunneling described in the framework of the orthodox theory. The first example of such cotunneling was the macroscopic quantum tunneling of the electric charge \((q-MQT)\) recently observed in 1D arrays of tunnel junctions. Another example is the low-frequency dissipation in single junctions in the Coulomb-blockade regime.

In this work we propose an example of the system in which charge transport can be completely due to cotunneling. An interesting aspect of this transport is that it qualitatively simulates the transport of solid-state microscopic excitons. In this respect the proposed system is the next one in a series of mesoscopic manmade systems that allow one to model the concepts of microscopic solid-state physics. Another system of this kind is the "artificial crystal" of Kouwenhoven et al., which exhibited the 1D band structure.

The proposed system consists of two 1D arrays of metal islands connected by small tunnel junctions and capacitively coupled as shown in Fig. 1. We assume that the tunneling between the arrays is negligible, so that we have two circuits that are independent galvanically but are coupled electrostatically. The electrostatics of the system is characterized by two capacitances: \( C_0 \) between the adjacent metal islands in the different arrays, and \( C \) between the islands connected by the junction (see Fig. 1). We are interested in the case \( C_0 \gg C \).

In this case the electrostatic energy \((\approx e^2/C_0)\) of the pair consisting of an electron in one of the islands and a hole in the adjacent island of another array is much smaller than the energy of the unpaired electron \((\approx e^2/C)\). Thus, it is energetically favorable to form such an "exciton" and significant energy is needed to separate the electron and the hole and destroy the exciton.

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**FIG. 1.** Schematic drawing and equivalent circuit of two electrostatically coupled 1D arrays of metal tunnel junctions. The dashed region denotes the insulator layers through which electrons can tunnel. Also shown is the charge distribution associated with an electron-hole pair ("exciton") on the adjacent electrodes of the two arrays. On the equivalent circuit the tunnel junctions are represented by crossed capacitances.
Excitons can enter the junction arrays and move along them even when the driving voltages applied to the system are much smaller than $e/C$ and the arrays are in the Coulomb-blockade state with respect to the single-electron processes. They move by hopping from site to site due to the two-electron cotunneling process. At first an electron tunnels virtually through the junction in one of the arrays, and increases the electrostatic energy. From the equivalent circuit of our system (shown in Fig. 1 for the specific case of three-junction arrays) one can be convinced that at $C_0 \gg C$ the energy $\epsilon$ of the resulting charge configuration is

$$\epsilon = (e^2/4C)(1 - 1/N),$$

where $N$ is the number of the junctions in one array. It can be viewed as the bonding energy of the exciton. The energy is decreased when the second electron tunnels through the coupled junction of another array in the direction opposite to that of the first electron. In other words, one can say that the hole tunnels in the same direction.) This tunneling restores the exciton charge configuration that is already shifted one site along the arrays.

The rate $\gamma$ of such a two-electron tunneling process is given by the same equations as the rate of the $q$-MOT in the double-junction system that was calculated in Refs. 3 and 7. In particular, in the limit of small temperatures $T$ and small energy gain $U$ associated with the two-electron tunneling process, $T, U \ll \epsilon$, we get from Eq. (10) of Ref. 7

$$\gamma = \frac{hU}{3\pi e^2 e^2 R^2} \frac{U^2 + (2\pi T)^2}{1 - \exp(-U/T)},$$

where $R$ is the junction resistance that we assume to be equal for all junctions.

In contrast to excitons in a real semiconductor crystal, in our "artificial crystal" the charges forming the "exciton" are spatially separated, and exciton transport leads to charge transport. Exciton hopping shifts one electron along the upper junction array and one electron along the lower array in the opposite direction. Thus, the currents in the circuits that contain the arrays are exactly equal in magnitude and opposite in direction. Moreover, the driving voltage for the exciton is the difference between the voltages across the two arrays. Hence, there is a possibility to establish the current against the voltage in one of the circuits. The energy needed to do this is supplied by the voltage in the other circuit.

These properties of the exciton transport do not depend on the number of the junctions in the arrays, and for driving voltages that are smaller than $e/C$ are independent of the voltage. However, the specific dynamics of exciton motion and the magnitude $I$ of the currents flowing in the arrays depend on both parameters.

At small driving voltages $V \approx e/C$ the correlations between excitons are important. Let us consider, for example, the system of three-junction arrays shown in Fig. 1. At low temperatures, $T \ll e^2/C_0$, for an exciton to enter the system the voltage should be larger than the threshold voltage $V_t \approx e/C$. (As usual, we assume that uncontrolled residual voltage differences between metallic islands of the system are negligible.) At $C_0 \gg C$ the driving voltage for the excitons drops completely across the edge junctions. Hence, at voltages $V$ slightly larger than $V_t$, $V - V_t \ll V_t$, the picture of the exciton motion through the system is as follows.

An exciton jumps into the system through one of the edges and waits on the site until the exciton of opposite sign (with reversed position of electron and hole) enters the system through another edge. Then one of them hops across the middle pair of junctions and the excitons annihilate. Since the different acts of exciton hopping are uncorrelated, the dynamics of exciton motion can be described quantitatively by the conventional master equation for probabilities $\sigma(n_1, n_2)$ to find $n_1$ excitons on the $i$th site. Solving the simple master equation that corresponds to the qualitative picture of exciton hopping described above, one can get the steady-state current $I$ flowing through the three-junction arrays at small driving voltages:

$$I = \frac{\hbar C^2(V-V_t)}{\pi R^2 e^4} \left(\frac{V-V_t}{2} + (4\pi T/e)^2\right)^2 \left[1 - \exp\left(-e(V-V_t)/2T\right)\right].$$

In the opposite limit, when the voltage and/or temperature is relatively large, $e^2/C_0 \ll eV, T \ll \epsilon$, the characteristic number of excitons on each site of the system is also large, $|n_i| \gg 1$, and the correlations between excitons are suppressed. In this case one can neglect both the discreteness of $n_i$ and $n_i$ fluctuations and reduce the master equation for $\sigma(n_1, \ldots, n_{N-1})$ to the equation for the average potential $\varphi_i$ between two metal islands of the $i$th site, $\varphi_i = e(n_i)/C_0$ ($i = 1, \ldots, N - 1$):

$$\varphi_i = (I_{i-1} - I_i)/C_0, \quad I_i = e(\gamma_{i+1} - \gamma_{i+1}).$$

Here $\gamma_{i+1}$ is the rate of the exciton hopping from the site $i$ to the site $i+1$, which is given by Eq. (2) with $U = e(\varphi_i - \varphi_{i+1})$, and $\varphi_0$ and $\varphi_{N+1}$ are the potential differences between, respectively, left and right external electrodes. At $T \gg e(\varphi_i - \varphi_{i+1})$, Eq. (4) is the discrete version of the diffusion equation with the diffusion constant $D$:

$$\varphi_i = D(\gamma_{i+1} + \gamma_{i-1} - 2\gamma_i),$$

$$D = (4\pi \hbar e^2 R^2 C_0^2)T/e^2.$$

Equation (4) shows that in the stationary state the potentials $\varphi_i$ change linearly along the arrays, $\varphi_i - \varphi_{i+1} = \text{const} = (V_u - V_d)/N$, where $V_u, V_d$ are the voltages across the upper and lower arrays. Thus, the excitons are driven by the difference of the voltages across the arrays, and the current $I$ through the arrays is $e \gamma(U)$.
\[ I = \frac{4hC^2V}{\pi R^2e^4} \left[ \frac{V}{e} \right]^2 + \left( \frac{2\pi T}{e} \right)^2 \]  

(6)

The dc I-V curve of this system for arbitrary voltages and \( T = 0 \) in the regime of the exciton transport is shown in Fig. 2. At small and large voltages this curve reaches the asymptotes (3) and (6), respectively. Deviations from the small-voltage asymptote at very small currents, which are visible on this scale, result from the fact that the \( C/C_0 \) ratio is finite. Note that the large-voltage asymptote is reached only on the log-log scale, since at \( V \ll \delta \), Eq. (6) gives correctly only the leading (in voltage) term that is proportional to \( V^2 \).

At finite \( C/C_0 \) ratios exciton transport can coexist with single-electron tunneling. For the three-junction arrays shown in Fig. 1 the zero-temperature boundaries between different regimes are displayed in Fig. 3. At small driving voltages \( V < e/C_0 \) the single-electron and exciton transport are suppressed. At some larger voltages only exciton transport becomes possible. For \( C_0 \gg C \) the boundaries of the parameter region for pure exciton transport are \( V = e/C_0 \) and \( V = e/4C \). At larger voltages \( e/4C < V < e/2C \) for \( C_0 \gg C \) single electrons cannot tunnel through the array, but the exciton motion can create charge configurations that make single-electron tunneling possible. Since at \( T = 0 \) the number \( n \) of excitons on the sites of the array changes discretely with increasing voltage, the boundary between the pure exciton and combined transport regions exhibits steps at \( VC_0/e \equiv 1, 3, 5, \ldots \), where \( n \) is increased by 1. Finally, at \( V > e/2C \) single electrons can tunnel along the array and charge transport is governed by single-electron tunneling.

In principle, the correlated single-electron tunneling can simulate to some extent the cotunneling responsible for the exciton transport. The tunneling of a single electron along one of the junction arrays drives the hole along the coupled array. However, in this case the electron and the hole are not completely bound together as they are in the exciton, so that the currents in the two arrays are not strictly equal. Numerical simulations of single-electron tunneling in the three-junction arrays (Fig. 1) confirm that the current \( I_u \) in the upper circuit induces a reverse current \( -I_d \) in the lower circuit, but the relative magnitude of these currents, \( I_u/I_d \), is rather small, and rapidly decreases with increasing voltage; see Fig. 4.

It means that in the regime of exciton transport thermally activated single-electron tunneling events decrease the accuracy of the equality of the currents in the two arrays. The relative magnitude of the single-electron contribution to the deviations from this equality can be roughly estimated as \( \exp[-(e-eV)/T] \). Another
source of inaccuracy is the above-mentioned $q$-MQT processes—quantum electron tunneling through the whole array. The rate of this tunneling is proportional to $(R_0/R)^N$, so that for the three-junction arrays the relative magnitude of the $q$-MQT currents can be estimated as $R_0/R$.

Before concluding, we discuss briefly the possibility of experimental realization of the proposed system with electrostatically coupled junction arrays (Fig. 1). This system differs from the conventional systems of submicron tunnel junctions only in the large interarray capacitance $C_0 \gg C$. One of the possible ways to fabricate these capacitances is to connect the adjacent electrodes of the arrays by additional metal strips evaporated over them. In this way one can make the ratio $C/C_0$ as small as 0.01, while keeping the junction capacitance $C$ (and the self-capacitances of the electrodes) close to the typical values $10^{-15} - 10^{-16}$ F. Thus, a slight modification of the existing fabrication technique should provide the possibility to observe the exciton transport considered above.

In conclusion, we have proposed a new system that should exhibit an unusual charge transport associated with the quantum tunneling of electron-hole pairs. The system is experimentally feasible and might have practical applications.

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