Thermoelectric phenomena in a one-dimensional diffusive quantum medium

I. N. Volovichev and D. V. Kadygrob
O. Ya. Usikov Institute for Radiophysics and Electronics, National Academy of Sciences of Ukraine, 12 Ac. Proscura Kharkov 61085, Ukraine

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The quantum diffusion in a one-dimensional lattice in a tunneling regime under the influence of spatially inhomogeneous Gaussian noise is studied theoretically. It is shown that the thermopower appears at an arbitrary value of the tunneling coupling. In the strong tunneling regime at an extremely high temperature, when the drift component of the charge transport can be neglected, the thermoelectric effect takes place only in the presence of a constant external electric field. The Seebeck and Peltier coefficients for the closed-circuit mode are obtained for a one-dimensional quantum diffusion medium in the tunneling regime. In the strong tunneling regime, both the thermopower and the Peltier effect are essentially nonlinear phenomena.

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I. INTRODUCTION

A stochastic potential being applied to a quantum one-dimensional (1D) system reveals a wide range of physical effects. An essential feature of quantum transport in a disordered 1D structure (e.g., a nanowire) is the phenomenon of the Anderson localization [1], preventing the charge transport through such a system. In many papers, the effect of a dynamic stochastic potential on the dynamics of a quantum system [2,3], in particular, on the ballistic quantum transport and charge transfer in 1D metallic systems [4] was studied. Among other things, the electron localization and delocalization processes were considered [4–6]. It is found that the presence of fluctuating potential in such a system (for example, at nonzero temperature due to the interaction with phonons) can break localization, giving rise to processes known as quantum diffusion [7–9] and superdiffusion [10,11] in inhomogeneous 1D systems. In the limiting case of quickly fluctuating potential and, therefore, lack of coherence, quantum transport in the localized system can be treated as classical hopping dynamics [6].

One of the simplest models of a 1D system, convenient for studying quantum diffusion, is an infinite linear chain of sites, each interacting only with its closest neighbors. In contrast to the case of the Anderson localization, the electron transport along the chain is assumed to be incoherent. The probabilities of the tunneling transition in such a system were intensively studied [12,13]. In Ref. [14] it is shown within the framework of the tight-binding model that, in a linear chain of weakly coupled atoms subjected to Gaussian noise, the quantum diffusion of electrons occurs and the diffusion coefficient is derived. In Ref. [15] this result is generalized to an arbitrary tunneling coupling within the Landau-Zener model. In particular, it was shown that, in the strong tunneling limit, the electron transport along the chain is pure diffusive by its nature, and there is no drift component. On the contrary, applying an external electric field suppresses the quantum diffusion (decreases the diffusion coefficient) and strengthens the electron localization.

When studying the effect of noise on the transport properties of the 1D chain, the noise parameters are widely considered to be spatially homogeneous. Here, we study the effect of spatial inhomogeneity on quantum diffusion in the infinite 1D linear chain of sites. The physical mechanisms of thermoelectric effects in 1D quantum diffusion systems at high temperatures are considered. Noise inhomogeneity can represent the spatial inhomogeneity of the temperature, allowing the study of the quantum thermodiffusion and thermoelectric phenomena in such a simple 1D quantum system with no ballistic charge transport.

In this paper, grounding on the model of a 1D diffusion medium proposed in [15], we investigate the electric current that flows in a closed circuit due to the spatial inhomogeneity of the amplitude of the noise potential correlation function and derive the Seebeck and Peltier coefficients.

II. STATEMENT OF THE PROBLEM

Let us consider the generation of the thermoelectric current in the 1D linear chain that is described by the Schrödinger equation with the following time-dependent Hamiltonian in the second quantization form:

$$H = \sum_j \Gamma\left(a_j^\dagger a_{j+1} + a_{j+1}^\dagger a_j\right) + \chi_j(t) a_j^\dagger a_j, \qquad (1)$$

where Γ is the tunneling constant and $a^\dagger{}_j$ and a_j are respectively the creation and annihilation operators in the jth site; $\chi_j(t)$ represents the fluctuations allowing diffusion in the system. We assume that $\chi_j(t)$ is the Gaussian noise being uncorrelated between nodes with time correlation law $\langle \chi_i(t')\chi_j(t+t')\rangle = \delta_{ij}C(t)$. The decay time of $C(t)$ is the correlation time τ; the typical magnitude of fluctuations is represented by W, which is defined as $W^2 = C(0)$; the characteristic noise velocity is defined as $v = cW/\tau$ with dimensionless constant $c > 1$.

The presence of noise in the Hamiltonian provides permanent repetition of the intersection of energy levels of

neighboring nodes, causing diabatic tunnel transitions. We consider all successive acts of tunneling between nodes incoherent and completely independent. We also consider the ballistic motion of the electron and its jumps over the lattice node (i.e., $j \to j \pm 2$) to be impossible.

According to the Landau-Zener theory [16,17], in the case under consideration, the transition probability p is given by [15]

$$p = 1 - \exp\left(-\frac{2\pi\Gamma^2}{\hbar v_C}\right), \tag{2}$$

where v_C is the energy level crossing velocity.

Obtaining kinetic coefficients involves averaging over v_C. It requires the distribution of v_C to be correctly defined. The noise sequences $\chi_j(t)$ must be differentiable to meet this requirement. Following [15], it can be obtained as a solution of a stochastic equation

$$m\ddot{\chi} + \eta\dot{\chi} + k\chi = \xi(t), \tag{3}$$

representing a harmonic oscillator excited by Gaussian white noise $\xi(t)$ with $\langle\xi(t)\xi(t')\rangle = \delta(t-t')$. The oscillator parameters in Eq. (3) are related to the desired parameters of the $\chi(t)$ distribution as follows [15]:

$$m = \frac{\tau^2}{Wc\tilde{c}}, \quad \eta = \frac{\tau\tilde{c}}{Wc}, \quad k = \frac{c}{W\tilde{c}}, \quad \tilde{c} = \sqrt{2(c^2+1)}.$$

To apply the Landau-Zener approach, the following conditions must be met. First, the fluctuation frequency should be small enough, so the crossing event duration should be sufficiently long. Second, adjacent energy levels should not affect the transitions by the Landau-Zener mechanism. Last, the inconstancy of the energy level crossing velocity in the tunneling time interval can be neglected. To meet these requirements of the model, the noise parameters must satisfy the following criteria, which are generalizations of ones derived in [15] for the spatially uniform noise:

$$\min(W_j) \gg c\Gamma, \quad \min(W_j) \gg \hbar c^3/\tau.$$

It should be noted that Eq. (2) might be incorrect in the "slow" Landau-Zener because of the Langevin noise in potential fluctuations. However, this issue may be neglected in the strong tunneling mode when the above conditions are met, and therefore the transition probability is close to unity.

We also assume that the spatial inhomogeneity of noise is small enough:

$$W_j = W_0 + j\Delta W, \quad \Delta W/W_0 \ll N^{-1}, \tag{4}$$

where N is the number of sites in the 1D lattice under consideration.

The Schrödinger equation for the Hamiltonian in Eq. (1) can be written down in terms of the wave function amplitudes of the nodes A_j as follows:

$$\imath\hbar\frac{dA_j}{dt} = \Gamma(A_{j+1} + A_{j-1}) + \chi_j(t)A_j. \tag{5}$$

Solving it by successive approximations on the small parameter Γ/W_0, one gets the following master equation for the probability $P_j(t) = |A_j|^2$ to find an electron in the jth site:

$$\frac{dP_j}{dt} = D[P_{j+1} + P_{j-1} - 2P_j] + D_T[P_{j+1} - P_{j-1}]\Delta W, \tag{6}$$

where $D = 2\Gamma^2\int_0^\infty C_\phi^2(t)dt$ is the diffusion coefficient, $C_\phi = \langle e^{-\imath\phi(t)}\rangle$, and $\phi(t) = \int_0^t \chi_0(t')dt'$.

Probabilities P_j in Eq. (5) should be considered as averaged probabilities over the ensemble of realizations of the noise sequences $\chi_j(t)$. As far as we consider the short circuit mode, there is no electric potential in Eq. (6). In the general case (open circuit or finite value of the resistance of the external load), the electric potential appears in the master equation. Below, it will be discussed in more detail when considering the strong tunneling coupling case.

When solving Eq. (5), two key assumptions are used. First, all terms containing product $(A_j^0)^*A_{j+1}^0$ (here A_j^0 is the zero-order solution in jth node) vanish when averaging over the ensemble. Second, diffusion is assumed to be a much slower process than dephasing. The latter assumption allows separating scales of the probability change rate and the dephasing rate.

The thermodiffusion coefficient D_T in Eq. (6) is defined as $D_T = \partial D/\partial W$.

III. RESULTS AND DISCUSSION

Identifying the amplitude of the noise correlation function with the temperature $W_j \sim k_BT$ (k_B is the Boltzmann constant; T is the temperature), we can refer to the diffusion processes under the influence of spatially inhomogeneous noise as to the thermodiffusion of charge carriers, being a physical origin of thermoelectric effects in the quantum diffusive medium. In the considered approximation, all thermoelectric effects result from quantum thermodiffusion. In turn, the thermodiffusion coefficient D_T is entirely defined by the functional dependence of the diffusion coefficient D on the noise magnitude W. Therefore, to calculate the value of thermoelectric parameters of the medium (the Seebeck and Peltier coefficients), it is necessary and sufficient to find the diffusion coefficient D. Analytical expressions for the diffusion coefficient for the model under consideration were obtained in [15]. Based on them, we obtain explicit expressions for the Seebeck coefficients in two opposite limiting cases when the expressions for $D(W)$ have the simplest form.

If the coupling is small enough $\Gamma^2\tau \ll \hbar cW_0$, the diffusion coefficient is $D = \sqrt{\pi}\Gamma^2/W$ [15]. Thus the thermodiffusion coefficient D_T is equal to

$$D_T = -D/W_0. \tag{7}$$

In the opposite limit case $\Gamma^2\tau \gg \hbar cW_0$, according to [15] the diffusion coefficient is independent of the noise magnitude in the node: $D = c/\pi\tau$. It means that, in the case of extremely strong tunneling, there is no thermodiffusion, $D_T = 0$. However, the situation changes if the 1D lattice under consideration is exposed to a constant uniform electric field E applied lengthwise to the chain. As shown in [15], in the case of the strong tunneling regime, the electric field suppresses

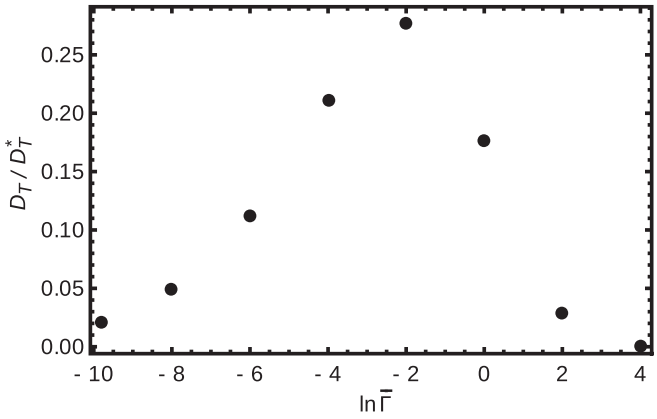

FIG. 1. Dependence of the normalized thermodiffusion coefficient on the tunneling constant and noise parameters, $D_T^* = 2c/\pi k_B T \tau$, and $\bar{\Gamma} = \pi^2 \Gamma^4 \tau^2 / 4c^2 W_0^2$.

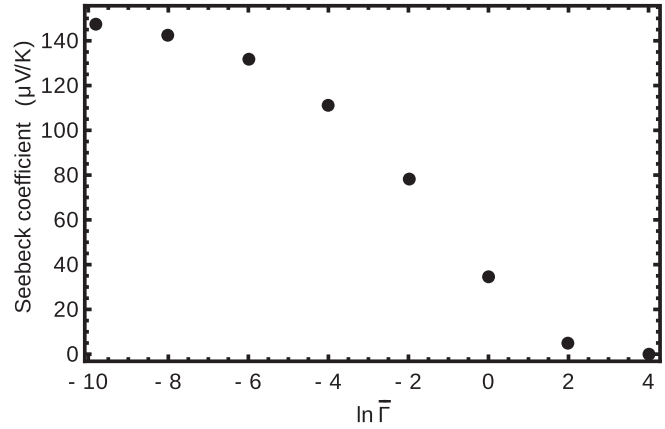

FIG. 2. Dependence of the Seebeck coefficient on the tunneling constant and noise parameters, $\bar{\Gamma} = \pi^2 \Gamma^4 \tau^2 / 4c^2 W_0^2$.

quantum diffusion: $D = D_0 e^{-e^2 \Delta V^2 / 4W_0^2}$, where D_0 is the diffusion coefficient without the electric field, $\Delta V = El$ is the difference of electric potentials in two neighboring nodes, and l is the lattice period.

Thus, in the case of the extremely strong tunneling regime in a constant electric field, the thermodiffusion is nonzero and is determined by the thermodiffusion coefficient

$$D_T = \frac{e^2 \Delta V^2}{2W_0^3} D. \tag{8}$$

Our results of numerical calculations of D_T for some intermediate values of the tunneling coupling are shown in Fig. 1.

As can be seen from Fig. 1, in the limiting cases of weak $\Gamma = 0$ and strong $\Gamma \to \infty$ tunneling coupling, there is no thermodiffusion. The physical reason why the thermodiffusion vanishes in these limiting cases is apparent. As follows from Eq. (2), at $\Gamma = 0$, there is no exchange of electrons between lattice sites. It means the disappearance of both diffusion and thermodiffusion in this limiting case. In the opposite limiting case $\Gamma \to \infty$, an electron passes with a probability of 1 to another lattice site. A random walk takes place and the diffusion coefficient remains nonzero. In this case, the tunneling probability does not depend on the noise magnitude at the node. Therefore, the diffusion coefficient is independent of the noise magnitude and the thermodiffusion coefficient equals zero. There is no physical reason for forming a directed flux of charge carriers in this limiting case, as the tunneling probability at each lattice node to any closest neighbor is the same.

Thus the thermodiffusion manifests itself to the maximum extent at a particular intermediate value of the tunneling constant. Our numerical calculation shows that the optimal value of the tunneling constant at which the thermodiffusion coefficient reaches its maximum is approximately equal to

$$\Gamma^* \approx \frac{1}{\pi} \sqrt{\frac{2W_0 c}{\tau}}.$$

The thermoelectric current arising in the system under consideration can be written in the usual form $j = -\sigma \alpha_T \nabla T$ [18] (here $\sigma = e\mu n$ is the conductivity of the medium, e is the elementary charge, μ is the electron mobility, and n is the electron density) if we consider the Seebeck coefficient α_T associated with the thermodiffusion. Indeed, the last term in Eq. (6) corresponds to the electric current $j = ek_B D_T n \nabla T$. Using the Einstein relation for the diffusion coefficient and the charge carrier mobility $D = \mu k_B T / e$, we obtain the following expressions for the Seebeck coefficient of the quantum diffusion medium under consideration:

$$\alpha_T = -\frac{k_B^2 T}{e} \frac{D_T}{D}. \tag{9}$$

In the case of weak tunneling coupling ($\Gamma^2 \tau \ll \hbar c W_0$), the Seebeck coefficient is $-k_B^2 T / eW_0$. Or, assuming $W_0 = k_B T$, we get the universal constant Seebeck coefficient depending on no medium parameter:

$$\alpha_T = -\frac{k_B}{e}. \tag{10}$$

In the opposite case of the strong tunneling regime ($\Gamma^2 \tau \gg \hbar c W_0$), the Seebeck coefficient can be formally written down as

$$\alpha_T = \frac{el^2 E^2}{2k_B T^2}. \tag{11}$$

According to Eq. (11), in the short circuit mode, the thermoelectric current vanishes because $E = 0$ in this case. For a finite resistance of the load, the thermoelectric effect takes place in the strong tunneling mode. However, it can be described by the Seebeck coefficient in the form as in Eq. (11) only formally, as the latter depends strongly on the applied electric field and, thus, on the load resistance and the thermoelectric current in the closed circuit.

Our numerical calculation results of the Seebeck coefficient for various values of the tunneling constant are depicted in Fig. 2.

Even in the limiting case of isolated lattice sites, the Seebeck coefficient remains nonzero, despite the disappearance of thermodiffusion, since in this case, the diffusion

coefficient also vanishes. Physically, monotonic decrease in the value of the Seebeck coefficient with the strengthening of the tunneling coupling is explained by the fact that, according to Eq. (2), the asymmetry of the tunneling probability monotonically decreases with increase of Γ.

It is interesting to compare the results above with thermoelectric properties of other physical models of solids with localized states and diffusion of charge carriers, studied by various methods. Among the closest models, there are 1D disordered conductors with metallic conductivity, classical disordered and amorphous conductors with hopping conductivity, and molecules of organic semiconductor polymers.

In the case of weak tunneling, the thermopower defined by Eq. (10) does not depend on the temperature and parameters of the medium. A disordered lattice of two-level sites with a nondegenerate electron gas at a sufficiently high temperature ($k_BT > \varepsilon_g$, where ε_g is the band gap) has similar properties [19]. Comparison of the model under consideration with organic semiconductor polymers is somewhat tricky because both the thermopower value and its temperature dependence rely on the chosen model for the density of states [20–24].

In the limiting case of strong tunneling, the thermoelectric properties of the considered model differ significantly from those described in the literature. The physical reason for the difference between our approach's results and other hopping transport models associated with the thermally activated transfer of electrons over potential barriers (e.g., the Miller-Abrahams approach [25,26]) is that we consider electrons only tunneling through barriers of infinite height.

Nevertheless, the trend common with many other models remains: the Seebeck coefficient decreases with increasing temperature if the system's conductivity decreases with increasing temperature [4,6]. In our opinion, some uniqueness of thermoelectric properties of the presented model in the strong tunneling case, compared to related models, is due to two points. First, in the present work, the thermoelectric parameters of the diffusion medium are obtained in the closed-circuit mode, while the Seebeck effect is usually studied in the open circuit mode [18]. Secondly, in the limit of strong tunneling, the medium under consideration is an insulator. The thermoelectric properties of weakly conductive media have been studied much less intensively since they are of no interest from the standpoint of applications as a thermal converter.

In Ref. [15], the physical reason for the vanishing conductivity in the strong tunneling limit has been revealed. The same physical reason suppresses the thermoelectric current in the problem under consideration. It can be formulated as follows. In the strong tunneling limit, the overbarrier hopping is negligible, electrons tunnel through the barrier, and the tunneling rate is determined mainly by the energy level change velocity rather than their position. The electric potential does not affect this velocity. The difference in the tunneling probabilities to left and right neighboring cities, which determines the drift current, vanishes. However, due to the Einstein relations, the diffusion rate remains nonzero, representing the high-temperature limit of a medium with vanishing conductivity. In the absence of the electric field, the thermal gradient does not break the equality of the tunnel probabilities in opposite directions, so thermal emf does not arise. In the uniform case, the external electric field just suppresses diffusion [15]. With the temperature gradient, the uniform external electric field makes the diffusion coefficient spatially nonuniform, giving rise to the thermoelectric current in the closed circuit.

Following the Onsager principle [27], the existence of thermal diffusion in the system under consideration means that the inverse process exists—the Peltier effect.

Using Onsager's relations [18,27], based on Eqs. (10) and (11), we can write down the Peltier coefficients for a 1D quantum lattice with the pure diffusive transport of electrons:

$$\Pi_T = \begin{cases} -\dfrac{k_BT}{e}, & \Gamma^2\tau \ll \hbar c W_0, \\ \dfrac{el^2E^2}{2k_BT}, & \Gamma^2\tau \gg \hbar c W_0. \end{cases} \tag{12}$$

In the strong tunneling regime, the Peltier effect, like the thermopower, is essentially nonlinear. Therefore, the Peltier coefficient introduced in Eq. (12) in the limiting case of the strong tunneling coupling should be regarded formally.

It should be emphasized that the thermoelectric effect studied in this paper is related to the peculiarities of quantum diffusion in the spatially uniform 1D system caused by applying spatially nonuniform stochastic potential. The presented analysis is obtained for the initial spatially uniform distribution of charge carriers at the lattice nodes. A nonuniform distribution of electrons is formed when the sample is connected to the external circuit, giving rise to diffusion and drift fluxes which tend to cancel out the initial thermodiffusion flux. In addition, in a closed circuit, along with the considered mechanism, other physical processes at the contact of the 1D lattice with the electrodes may play an important role. The joint manifestation of these mechanisms forms the resulting thermoelectric effect, the magnitude of which may differ significantly from Eqs. (10) and (11).

IV. CONCLUSIONS

The above analysis shows that, in the infinite linear 1D chain of sites coupled by tunnel junctions, the spatial inhomogeneity of the random fluctuating potential leads to the quantum thermodiffusion, generating the electric current in the closed circuit. Thermopower strongly depends on the strength of the tunnel coupling between the lattice sites. In the strong tunneling regime, when the drift component of the current is negligible and the diffusion coefficient does not depend on the noise amplitude, the thermoelectric effect occurs only in the presence of the electric field. In this case, the Seebeck coefficient quadratically depends on the electric field and increases with decreasing the average temperature of the system. In the case of a weak tunneling regime, the Seebeck coefficient does not depend on the temperature and medium parameters, being of the order of 10^{-4} V/K.

The ability of the spatial noise inhomogeneity to result in the appearance of a directed diffusion electron flux along the chain of sites bonded by tunnel junctions proves not only the existence of a direct thermoelectric effect (thermopower) but also means the possibility of manifestation of the inverse thermoelectric effect—the Peltier effect.

If the tunneling coupling is weak in the system under consideration, the Seebeck and Peltier coefficients are

universal constants not depending on the material properties. The Peltier coefficient depends on the temperature linearly. In the strong tunneling regime, both the Seebeck effect and the Peltier effect are essentially nonlinear effects and exist only in the presence of the electric field along the length of the 1D system.

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