Single-particle distribution function of a quantum dot system at finite temperature*

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In the present paper, we shall rigorously re-establish the result of the single-particle function of a quantum dot system at finite temperature. Unlike the proof given in our previous work (*Phys. Rev.* B **74** 195414 (2006)), we take a different approach, which does not exploit the explicit expression of the Gibbs distribution function. Instead, we only assume that the statistical distribution function of the quantum dot system is thermodynamically stable. As a result, we are able to show clearly that the electronic structure in the quantum dot system is completely determined by its thermodynamic stability. Furthermore, the weaker requirements on the statistical distribution function also make it possible to apply the same method to the quantum dot systems in non-equilibrium states.

Keywords: quantum dot systems, electron distribution function, rigorous results

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1. Introduction

As is well known, equilibrium statistical mechanics has been very successful in explaining the thermodynamic properties of macroscopic materials.[1] Starting from the microscopic Hamiltonian of a given many-body system, statistical mechanics allows us to derive its thermodynamic quantities, such as the free energy density, the specific heat, and the magnetic susceptibility, in a systematic manner. Perhaps, the greatest achievement of the equilibrium statistical mechanics is understanding the critical phenomena by using the renormalization group method.[2]

On the other hand, the simple and beautiful mathematical structure of statistical mechanics also provides some deep questions for mathematical physicists to ponder. For instance, it is easy to show that the ground-state energy of a many-body system must be an extensive quantity, which is proportional to N, the total number of particles in the system.[1] Otherwise, the free energy density of the system does not exist in the thermodynamic limit. In other words, it becomes unstable. Starting from this fact, Dyson and Lenard proved that a neutral solid is stable.[3,4] Later, their results were simplified by Lieb and Thirring.[5] Furthermore, those authors also showed that if electrons in the solid were bosons, then the system cannot be stable. Therefore, Heisenberg's uncertainty principle alone is not sufficient to guaranty the stability of matter as one would have believed before. More precisely, it is found that Pauli's exclusive principle also plays an essential role in achieving the stability.[6]

In a previous work,[7] we studied the restriction imposed on the microscopic single-particle function of a quantum dot system by another characteristic of the equilibrium statistical mechanics, i.e., the convexity of the Gibbs distribution function $\mathrm{e}^{-\beta E}$. We showed that the single-particle function is a non-increasing function with respect to the on-dot single-particle energy levels at finite temperature. Considering that the convexity of the Gibbs function is closely related to the stability of a many-body system in equilibrium,[8] we concluded that the monotonic property of the microscopic single-particle function is actually dictated by the stability requirement of the quantum dot system.

However, since we exploited heavily the explicit expression of the Gibbs function in our previous work, the connection between the stability of the quantum dot system and the non-increasing property of its on-dot single-particle function was only established in an indirect way. That is certainly a shortcoming. Naturally, we would like to find a more transparent approach to re-establish the result, if it is possible. Secondly, our previous proof cannot be extended to the case in which a small external voltage is applied to

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the conducting leads of the quantum dot system and hence the Gibbs function is no longer its distribution function.

In order to remedy these problems, we would like to re-establish our previous results by using a different approach to that followed in the present work. In the following, we shall only require that the distribution function of the quantum dot is thermodynamically stable, i.e., it is a positive and non-increasing function of the total energy of the system. Obviously, the requirement is much weaker and is certainly satisfied by the Gibbs function. Furthermore, since the explicit form of the distribution function is not needed by the present approach, it should be also applicable to derive similar conclusions for the quantum dot system in slightly non-equilibrium states.

This paper is organized as follows. In Section 2, we introduce the Hamiltonian of the quantum dot system and the definition of its single-particle function. In Section 3, we state and prove our main result for the single-particle function of the quantum dot. We show that it is a non-increasing function with respect to the single-particle energy levels. Finally, in Section 4, we summarize our results.

2. General Hamiltonian of quantum dot system

According to the standard textbook,[9] the Hamiltonian of a quantum dot system can be written as

$$\begin{aligned}\hat{H} = & \sum_{k,\,\sigma,\,\alpha}(\epsilon_{k\alpha}-\mu_\alpha)\hat{c}^\dagger_{k\sigma\alpha}\hat{c}_{k\sigma\alpha} \\ & + \sum_m\sum_\sigma \epsilon_m \hat{d}^\dagger_{m\sigma}\hat{d}_{m\sigma} + U\sum_m \hat{n}^d_{m\uparrow}\hat{n}^d_{m\downarrow} \\ & + V\sum_{m\neq m'}\left(\hat{n}^d_{m\uparrow}+\hat{n}^d_{m\downarrow}\right)\left(\hat{n}^d_{m'\uparrow}+\hat{n}^d_{m'\downarrow}\right) \\ & + \sum_\alpha t_\alpha \sum_{m,\,k,\,\sigma}\left(\hat{c}^\dagger_{k\sigma\alpha}\hat{d}_{m\sigma}+\hat{d}^\dagger_{m\sigma}\hat{c}_{k\sigma\alpha}\right), \quad (1)\end{aligned}$$

where $\alpha = \mathrm{R}$, L represents the right and the left conducting leads, respectively. They are connected to the quantum dot via electron tunneling, and t_α denotes the corresponding tunneling amplitude. The $\hat{c}^\dagger_{k\sigma\alpha}$ ($\hat{c}_{k\sigma\alpha}$) is the creation (annihilation) fermion operator for the conduction electron in lead α with spin $\sigma = \uparrow, \downarrow$ and single-particle energy $\epsilon_{k\alpha}$. Furthermore, $\hat{d}^\dagger_{m\sigma}$ and $\hat{d}_{m\sigma}$ denote the creation and the annihilation operators of electron with spin $\sigma = \uparrow, \downarrow$ and energy ϵ_m on the quantum dot. The positive parameters U and V represent the repulsive Coulomb interaction of electrons in the same and different levels, respectively. Naturally, these parameters may be level-dependent in real materials. However, for a quantum dot, only a few on-dot single-particle energy levels are actually active. Consequently, we can simply ignore the dependences of these parameters on the single-particle energy $\{\epsilon_m\}$. In experiments, an external chemical potential μ_α is imposed to conducting lead α. The difference $V_\mathrm{B} \equiv (\mu_\mathrm{R} - \mu_\mathrm{L})/e$ is defined to be the external electric voltage (bias) applied to the system.

The total current through the quantum dot system can be calculated by using the Keldysh Green function method.[10] It is given by[9]

$$\begin{aligned}J = & \frac{\mathrm{i}e}{\hbar}\int\frac{\mathrm{d}\epsilon}{2\pi}\left(f_\mathrm{L}(\epsilon)-f_\mathrm{R}(\epsilon)\right) \\ & \times \mathrm{Tr}\left[\frac{\boldsymbol{\Gamma}_\mathrm{L}(\epsilon)\boldsymbol{\Gamma}_\mathrm{R}(\epsilon)}{\boldsymbol{\Gamma}_\mathrm{L}(\epsilon)+\boldsymbol{\Gamma}_\mathrm{R}(\epsilon)}\left(\boldsymbol{G}^\mathrm{r}_\sigma(\epsilon)-\boldsymbol{G}^\mathrm{a}_\sigma(\epsilon)\right)\right], \quad (2)\end{aligned}$$

where $\boldsymbol{G}^\mathrm{r}_\sigma(\epsilon)$ and $\boldsymbol{G}^\mathrm{a}_\sigma(\epsilon)$ are the Fourier transformed matrices of the retarded Green function

$$G^\mathrm{r}_{\sigma,mn}(t,\,t') \equiv -\mathrm{i}\theta(t-t')\langle\{\hat{d}_m(t),\,\hat{d}^\dagger_n(t')\}\rangle \quad (3)$$

and the advanced one, respectively. Moreover, the level-width matrices $\boldsymbol{\Gamma}_\mathrm{L}(\epsilon)$ and $\boldsymbol{\Gamma}_\mathrm{R}(\epsilon)$ are defined by $[\boldsymbol{\Gamma}_\alpha(\epsilon)]_{mn} = 2\pi|t_\alpha|^2\rho_\alpha(\epsilon)\delta_{mn}$. Here, $\rho_\alpha(\epsilon)$ is the electron density of states in conducting lead α, $f_\mathrm{L}(\epsilon)$ and $f_\mathrm{R}(\epsilon)$ denote the well-known Fermi distribution functions of electron in the corresponding leads. By its definition, current J is a function of the external voltage V_B. Experimentally, one is mainly interested in measuring the differential conductance $g(V_\mathrm{B}) = \mathrm{d}J(V_\mathrm{B})/\mathrm{d}V_\mathrm{B}$ of the system.[11,12]

Generally, it is often assumed that only one energy level on the dot is active, since its diameter is of the nanometer scale. Consequently, each matrix in Eq. (2) can be replaced by a complex number. Furthermore, we can apply the equation of motion method[9] to evaluate Green functions $G^\mathrm{r}_\sigma(\epsilon)$ and $G^\mathrm{a}_\sigma(\epsilon)$. Explicitly, the retarded Green function is given by

$$\begin{aligned}G^\mathrm{r}_\sigma(\epsilon) = & \frac{\langle\hat{n}^d_{\bar{\sigma}}\rangle}{\epsilon-\epsilon_0-U+\mathrm{i}\Gamma(\epsilon)/2} \\ & + \frac{1-\langle\hat{n}^d_{\bar{\sigma}}\rangle}{\epsilon-\epsilon_0+\mathrm{i}\Gamma(\epsilon)/2}, \quad (4)\end{aligned}$$

where $\Gamma(\epsilon) = \Gamma_\mathrm{L}(\epsilon)+\Gamma_\mathrm{R}(\epsilon)$, and $\langle\hat{n}^d_{\bar{\sigma}}\rangle$ is the electronic single-particle function of spin $\bar{\sigma} = -\sigma$ on the dot. At finite temperature T, this function is defined by

$$n^d_{\bar{\sigma}} \equiv \langle\hat{n}^d_{\bar{\sigma}}\rangle = \frac{\mathrm{Tr}\left[\hat{n}^d_{\bar{\sigma}}\exp(-\beta\hat{H})\right]}{\mathrm{Tr}\exp(-\beta\hat{H})}. \quad (5)$$

Therefore, we need to determine both $n^d_\uparrow$ and $n^d_\downarrow$ first before proceeding to the calculation of the differential conductance of the quantum dot system.

In practice, the single-particle functions $n^d_\uparrow$ and $n^d_\downarrow$ are calculated by numerical methods. For instance, $n^d_\uparrow$ is related to the lesser Green function $G^<_{d\uparrow}(t-t') \equiv \mathrm{i}\langle \hat{d}^\dagger_\uparrow(t')\hat{d}_\uparrow(t)\rangle$ by[9]

$$n^d_\uparrow = \langle \hat{n}^d_\uparrow \rangle = -\mathrm{i}\int \frac{\mathrm{d}\epsilon}{2\pi} G^<_{d\uparrow}(\epsilon). \tag{6}$$

And $G^<_{d\uparrow}(\epsilon)$ can be computed by either the non-crossing approximation[13−15] or the charge-conserving ansatz.[16,17] However, the errors caused by these approximations are unavoidable and difficult to estimate. Moreover, it is not easy to deal with the quantum dot system with more than two active on-dot levels. That is the reason why such systems are hardly discussed in the literature. Consequently, we would like to ask whether some rigorous restrictions can be imposed on the on-dot single-particle functions $n^d_\uparrow$ and $n^d_\downarrow$. Obviously, these restrictions can be used as reliable and independent tests to the results derived by either the non-crossing approximation or the charge-conserving ansatz. Furthermore, by rigorous investigations, we will reach a deeper understanding on the effects of the electronic correlations in these small structures of nanometer scale.

In Ref. [7], we established a rigorous result on the single-particle function of a quantum dot system with more than two active on-dot levels. In that case, the function depends explicitly on the single-particle energy level index m and is given by

$$n^d_m = \frac{\mathrm{Tr}\left[\left(\hat{n}^d_{m\uparrow} + \hat{n}^d_{m\downarrow}\right)\exp(-\beta\hat{H})\right]}{\mathrm{Tr}\exp(-\beta\hat{H})}. \tag{7}$$

For n^d_m, we were able to show that it is a non-increasing function of index m. In other words, inequality $n^d_{m'} \le n^d_m$ holds for any pair of on-dot energy levels $\epsilon_{m'} \ge \epsilon_m$. To prove that result, we used the fact that the Gibbs distribution operator $\exp(-\beta\hat{H})$, which describes a many-body system in equilibrium, is convex with respect to its variable, as we mentioned in the previous section. Consequently, the deep connection between the monotonic property of the single-particle function with respect to the single-particle energy levels and the thermodynamic stability of the quantum dot system cannot be as clearly seen as it should be. In the next section, we shall provide a different proof, which does not require the explicit expression of the distribution operator and hence remedies the above-mentioned problem.

3. Rigorous result on single-particle function

To begin with, we require that the thermodynamic properties of the quantum dot system can be described by a distribution operator $f(\hat{H})$, with $f(x)$ being a differentiable function with respect to its variable x. Obviously, when the external bias $V_\mathrm{B} \equiv (\mu_\mathrm{R} - \mu_\mathrm{L})/e$ imposed on the conducting leads is zero, and hence the system is in equilibrium, this requirement is satisfied. In this case, the distribution operator is explicitly given by the well-known Gibbs operator $f(\hat{H}) = \exp(-\beta\hat{H})$. However, with a finite external bias $V_\mathrm{B} \neq 0$ being imposed on the conducting leads, the quantum dot system is in a non-equilibrium state. Consequently, this requirement may not be satisfied. Fortunately, some authors argued that it should still hold if the external bias is not very large, although $f(\hat{H})$ is no longer represented by the Gibbs function in this case.[18,19]

Next, we require that the quantum dot system is stable in the following sense: its Hamiltonian has a finite ground-state energy $E_0 > -\infty$. This requirement may seem trivial. However, as is well known, a relativistic quantum mechanical system described by the Dirac equation does not satisfy it. In a previous work,[20] we showed that this requirement alone is sufficient to determine the non-increasing property of the single-particle function with respect to the on-dot energy level index m at $T = 0$. However, at finite temperature, two more conditions must be imposed on the distribution operator $f(\hat{H})$ of the quantum dot system to attain its thermodynamic limit. First, by its definition, $f(E_n) \ge 0$ should hold for any eigenvalue E_n of the Hamiltonian. In other words, the negative probability is excluded from our consideration. Secondly, $f(\hat{H})$ should be non-increasing with respect to the eigenvalues of $\hat{H}$, i.e., $f(E_1) \ge f(E_2)$ holds for any pair of eigenvalues $E_1 \le E_2$. Otherwise, the quantum dot system will tend to stay in a state with a higher total energy. In the stationary case, this is impossible from an experimental point of view.

With these preparations, we are now ready to state and prove our main result in the present work.

Theorem 1 Let n^d_m be the on-dot single-particle function defined by

$$n^d_m = \frac{\mathrm{Tr}\left[\left(\hat{n}^d_{m\uparrow} + \hat{n}^d_{m\downarrow}\right) f(\hat{H})\right]}{\mathrm{Tr}\, f(\hat{H})}, \tag{8}$$

where $f(\hat{H})$ is the distribution operator satisfying the above-mentioned stability conditions. Then, n_m^d is a non-increasing function with respect to the on-dot single-particle energy level index m, i.e., for any pair of on-dot energy levels $\epsilon_{m'} > \epsilon_m$, inequality $n_{m'}^d \leq n_m^d$ holds.

Before proceeding to the proof of Theorem 1, we would like to give a remark on the physical meaning of it. When temperature $T = 0$ and external bias $V_{\rm B} = 0$, the quantum dot system is in its equilibrium ground state Ψ_0. Assume that all the interactions in Hamiltonian (1) are temporarily turned off by setting $U = V = t = 0$, then, the central dot of the system will be isolated. In this case, the electrons on the dot become free fermions. Therefore, according to Pauli's exclusive principle, they will completely fill up the lower energy levels below the Fermi energy $E_{\rm F}$. Consequently, we find that $n_m^d = 2$ for $\epsilon_m \leq E_{\rm F}$ and $n_m^d = 0$ for $\epsilon_m > E_{\rm F}$. In this case, Theorem 1 is certainly true. However, after having turned on the interactions U, V, and t, the electrons in the energy levels below $E_{\rm F}$ will be partially removed and added to the higher energy levels above the Fermi energy. As a result, this monotonic property of the single-particle function may be violated. Theorem 1 tells us that this situation cannot occur if the quantum dot system is thermodynamically stable.

Now, we shall prove Theorem 1.

Proof To start with, we introduce an integral function

$$F(E) = \int_{E_0}^{E} f(x)\,{\rm d}x, \tag{9}$$

where E_0 is the lowest eigenvalue of Hamiltonian $\hat{H}$. Since $f(x)$ is assumed to be a positive, differentiable, and non-increasing function, we obtain immediately the relations

$$F'(E) = f(E) \geq 0, \quad F''(E) = f'(E) \leq 0 \tag{10}$$

by taking the derivatives of $F(E)$ with respect to its variable. In other words, $F(E)$ is a non-decreasing and concave function of variable E. For such a function, we have the following fact.

Proposition 1 Let E_1 and E_2 be two real numbers in interval (E_0, ∞). Then, inequality

$$\begin{aligned} & F(E_2) - F(E_1) - F'(E_1)(E_2 - E_1) \\ = \; & F(E_2) - F(E_1) - f(E_1)(E_2 - E_1) \leq 0 \end{aligned} \tag{11}$$

holds.

A proof of the above proposition can be found in any standard textbook on convex functions. For example, one can read page 25 of Ref. [21].

Keeping the above fact in mind, we shall now establish the following Klein-type inequality, which plays a fundamentally important role in studying quantum statistical mechanics.[1]

Proposition 2 Let $\hat{A}$ and $\hat{B}$ be two Hermitian matrices. Assume that their eigenvalues belong to the domain of a concave function $F(x)$, then, inequality

$$\begin{aligned} & {\rm Tr}\left(F(\hat{A}) - F(\hat{B}) - F'(\hat{B})(\hat{A} - \hat{B})\right) \\ = \; & {\rm Tr}\left(F(\hat{A}) - F(\hat{B}) - f(\hat{B})(\hat{A} - \hat{B})\right) \leq 0 \end{aligned} \tag{12}$$

holds.

Proof Following Ref. [1], we take an arbitrary complete set of orthonormal eigenvectors $\{\phi_i\}$ for Hermitian matrix $\hat{A}$ and another set $\{\psi_i\}$ for $\hat{B}$. Let $\{a_i\}$ and $\{b_i\}$ be the corresponding eigenvalues of these eigenvectors. Then, for each eigenvector ϕ_i of matrix $\hat{A}$, we have

$$\begin{aligned} & \left\langle \phi_i \left| F(\hat{A}) - F(\hat{B}) - f(\hat{B})(\hat{A} - \hat{B}) \right| \phi_i \right\rangle \\ = \; & F(a_i) - \sum_{j=1}^{N} |c_{ij}|^2 F(b_j) - \sum_{j=1}^{N} |c_{ij}|^2 f(b_j)(a_i - b_j) \\ = \; & \sum_{j=1}^{N} |c_{ij}|^2 \left[F(a_i) - F(b_j) - f(b_j)(a_i - b_j)\right] \\ \leq \; & 0. \end{aligned} \tag{13}$$

In deriving inequality (13), we insert identity decomposition $\hat{I} = \sum_{j=1}^{N} |\psi_j\rangle\langle\psi_j|$ in several places. The coefficient $c_{ij} = \langle\phi_i|\psi_i\rangle$ represents the overlapping between vectors ϕ_i and ψ_i. We also apply inequality (11) in the last step of the above derivation. Finally, we sum over index i and accomplish the proof of Proposition 2.

In order to prove Theorem 1 with Proposition 2, we take the following strategy. In the first step, we assume that the theorem is false. Starting from this assumption, we introduce two unitary transformations that map Hamiltonian $\hat{H}$ of the quantum dot system into a new form $\widetilde{\hat{H}}$. Then, we let $\hat{A} = \widetilde{\hat{H}}$ and $\hat{B} = \hat{H}$ and substitute them into the left-hand side of inequality (12). A straightforward calculation will reveal that this inequality is actually violated if our starting assumption is true. Therefore, the assumption must be wrong to make the final conclusions consistent. Then Theorem 1 is proved.

Let us first rewrite Hamiltonian (1) into a more

symmetric form. Actually, we have

$$\begin{aligned}\hat{H} = & \sum_{k,\,\sigma,\,\alpha}(\epsilon_{k\sigma}-\mu_\alpha)\hat{c}^\dagger_{k\sigma\alpha}\hat{c}_{k\sigma\alpha}+\sum_m\sum_\sigma \epsilon_m \hat{d}^\dagger_{m\sigma}\hat{d}_{m\sigma}\\ & +(U-2V)\sum_m \hat{n}^d_{m\uparrow}\hat{n}^d_{m\downarrow}-V\sum_m\sum_\sigma \hat{n}^d_{m\sigma}\\ & +V\left[\sum_m\left(\hat{n}^d_{m\uparrow}+\hat{n}^d_{m\downarrow}\right)\right]^2\\ & +\sum_\alpha t_\alpha \sum_{m,\,k,\,\sigma}\left[\hat{c}^\dagger_{k\sigma\alpha}\hat{d}_{m\sigma}+\hat{d}^\dagger_{m\sigma}\hat{c}_{k\sigma\alpha}\right]. \end{aligned} \tag{14}$$

Next, we assume that there are at least two single-particle energy levels $\epsilon_{m_1} > \epsilon_{m_2}$, for which inequality $n^d_{m_1} > n^d_{m_2}$ holds. In other words, Theorem 1 is not valid for these energy levels.

To show this assumption must be wrong, we define a unitary transformation $\hat{U}_1(m_1,\, m_2)$ by

$$\begin{aligned}\hat{U}_1(m_1,\, m_2) & \equiv \exp\left[\frac{\mathrm{i}\pi}{2}\left(\hat{d}^\dagger_{m_1\uparrow}\hat{d}_{m_2\uparrow}+\hat{d}^\dagger_{m_2\uparrow}\hat{d}_{m_1\uparrow}\right.\right.\\ & \left.\left.+\hat{d}^\dagger_{m_1\downarrow}\hat{d}_{m_2\downarrow}+\hat{d}^\dagger_{m_2\downarrow}\hat{d}_{m_1\downarrow}\right)\right]. \end{aligned} \tag{15}$$

By applying the Baker–Hausdorff formula

$$\begin{aligned} & \mathrm{e}^{\hat{A}}\hat{B}\,\mathrm{e}^{-\hat{A}}\\ & =\hat{B}+\left[\hat{A},\,\hat{B}\right]+\frac{1}{2!}\left[\hat{A},\,\left[\hat{A},\,\hat{B}\right]\right]+\cdots, \end{aligned} \tag{16}$$

we can easily show that electron operators $\hat{d}_{m_1\sigma}$ and $\hat{d}_{m_2\sigma}$ in Hamiltonian (14) are interchanged under this transformation. More precisely, we have

$$\begin{aligned}\hat{U}^\dagger_1(m_1,\, m_2)\hat{d}_{m_1\sigma}\hat{U}_1(m_1,\, m_2)=\mathrm{i}\hat{d}_{m_2\sigma},\\ \hat{U}^\dagger_1(m_1,\, m_2)\hat{d}_{m_2\sigma}\hat{U}_1(m_1,\, m_2)=\mathrm{i}\hat{d}_{m_1\sigma}. \end{aligned} \tag{17}$$

At the same time, the other fermion operators are intact. As a result, by applying $\hat{U}_1(m_1,\, m_2)$, we map Hamiltonian (14) into

$$\begin{aligned}\hat{H}' &= \hat{U}^\dagger_1\hat{H}\hat{U}_1\\ &= \sum_{k,\,\sigma,\,\alpha}(\epsilon_{k\sigma}-\mu)\hat{c}^\dagger_{k\sigma\alpha}\hat{c}_{k\sigma\alpha}+(U-2V)\sum_m \hat{n}^d_{m\uparrow}\hat{n}^d_{m\downarrow}-V\sum_m\sum_\sigma \hat{n}_{m\sigma}+V\left[\sum_m\left(\hat{n}^d_{m\uparrow}+\hat{n}^d_{m\downarrow}\right)\right]^2\\ &\quad +\sum_\alpha t_\alpha\sum_{k,\,\sigma}\sum_m{}'\left(\hat{c}^\dagger_{k\sigma\alpha}\hat{d}_{m\sigma}+\hat{d}^\dagger_{m\sigma}\hat{c}_{k\sigma\alpha}\right)+\left(\epsilon_{m_1}\hat{n}^d_{m_2\uparrow}+\epsilon_{m_1}\hat{n}^d_{m_2\downarrow}+\epsilon_{m_2}\hat{n}^d_{m_1\uparrow}+\epsilon_{m_2}\hat{n}^d_{m_1\downarrow}\right)\\ &\quad +\sum_m{}'\sum_\sigma \epsilon_m\hat{d}^\dagger_{m\sigma}\hat{d}_{m\sigma}+\sum_\alpha t_\alpha\sum_{k,\,\sigma}\left(\mathrm{i}\hat{c}^\dagger_{k\sigma\alpha}\hat{d}_{m_1\sigma}-\mathrm{i}\hat{d}^\dagger_{m_1\sigma}\hat{c}_{k\sigma\alpha}+\mathrm{i}\hat{c}^\dagger_{k\sigma\alpha}\hat{d}_{m_2\sigma}-\mathrm{i}\hat{d}^\dagger_{m_2\sigma}\hat{c}_{k\sigma\alpha}\right), \end{aligned} \tag{18}$$

where $\sum'_m$ represents the sum over the single-particle energy levels with both ϵ_{m_1} and ϵ_{m_2} being excluded.

We would like to emphasize that an additional phase factor i appears in the tunneling interactions of the new Hamiltonian. In order to eliminate this extra factor, we introduce another unitary transformation $\hat{U}_2$, which is given by

$$\hat{U}_2 \equiv \exp\left(\frac{\mathrm{i}\pi}{2}\sum_{k,\,\sigma,\,\alpha}\hat{n}^c_{k\sigma\alpha}+\frac{\mathrm{i}\pi}{2}\sum_m{}'\hat{n}^d_{m\sigma}\right). \tag{19}$$

Under this transformation, the fermion operators are changed according to the following rules:

$$\begin{aligned}&\hat{U}^\dagger_2\hat{c}_{k\sigma\alpha}\hat{U}_2=\mathrm{i}\hat{c}_{k\sigma\alpha},\\ &\hat{U}^\dagger_2\hat{c}^\dagger_{k\sigma\alpha}\hat{U}_2=-\mathrm{i}c^\dagger_{k\sigma\alpha},\\ &\hat{U}^\dagger_2\hat{d}_{m\sigma}\hat{U}_2=\mathrm{i}\hat{d}_{m\sigma},\\ &\hat{U}^\dagger_2\hat{d}^\dagger_{m\sigma}\hat{U}_2=-\mathrm{i}\hat{d}^\dagger_{m\sigma},\\ &\quad \text{for}\ \ m\neq m_1,\, m_2. \end{aligned} \tag{20}$$

Consequently, Hamiltonian $\hat{H}'$ is further mapped into

$$\begin{aligned}\widetilde{\hat{H}} &= \hat{U}^\dagger_2\hat{H}'\hat{U}_2=\left(\hat{U}_1(m_1,\, m_2)\hat{U}_2\right)^\dagger\hat{H}\left(\hat{U}_1(m_1,\, m_2)\hat{U}_2\right)\\ &= \sum_{k,\,\sigma,\,\alpha}(\epsilon_{k\sigma}-\mu)\hat{c}^\dagger_{k\sigma\alpha}\hat{c}_{k\sigma\alpha}+(U-2V)\sum_m \hat{n}^d_{m\uparrow}\hat{n}^d_{m\downarrow}-V\sum_m(\hat{n}_{m\uparrow}+\hat{n}_{m\downarrow})+V\left[\sum_m\left(\hat{n}^d_{m\uparrow}+\hat{n}^d_{m\downarrow}\right)\right]^2\\ &\quad +\sum_\alpha t_\alpha\sum_{m,\,k,\,\sigma}\left(\hat{c}^\dagger_{k\sigma\alpha}\hat{d}_{m\sigma}+\hat{d}^\dagger_{m\sigma}\hat{c}_{k\sigma\alpha}\right)+\sum_m{}'\sum_\sigma \epsilon_m\hat{d}^\dagger_{m\sigma}\hat{d}_{m\sigma}+\left(\epsilon_{m_1}\hat{n}^d_{m_2\uparrow}+\epsilon_{m_1}\hat{n}^d_{m_2\downarrow}+\epsilon_{m_2}\hat{n}^d_{m_1\uparrow}+\epsilon_{m_2}\hat{n}^d_{m_1\downarrow}\right) \end{aligned} \tag{21}$$

by $\hat{U}_2$. Obviously, the electron tunneling interactions are completely recovered in this Hamiltonian. By comparing $\widetilde{\hat{H}}$ with the original Hamiltonian $\hat{H}$, we find that the only difference between them is that operators $\hat{n}^d_{m_1}$ and $\hat{n}^d_{m_2}$ are interchanged in the sum over ϵ_m.

Now, we let $\hat{A} \equiv \widetilde{\hat{H}}$ and $\hat{B} \equiv \hat{H}$. As shown above, $\widetilde{\hat{H}}$ is unitarily equivalent to $\hat{H}$. Therefore, $F(\widetilde{\hat{H}})$ is also equivalent to $F(\hat{H})$. Consequently, they have the same spectrum. It leads to the following identity:

$$\mathrm{Tr}F(\hat{A}) = \mathrm{Tr}F(\widetilde{\hat{H}}) = \mathrm{Tr}F(\hat{H}) = \mathrm{Tr}F(\hat{B}). \quad (22)$$

Moreover, the difference between $\widetilde{\hat{H}}$ and $\hat{H}$ is given by

$$\begin{aligned} &\hat{A} - \hat{B} \\ &= \epsilon_{m_1}\hat{n}^d_{m_2\uparrow} + \epsilon_{m_1}\hat{n}^d_{m_2\downarrow} + \epsilon_{m_2}\hat{n}^d_{m_1\uparrow} + \epsilon_{m_2}\hat{n}^d_{m_1\downarrow} \\ &\quad - \epsilon_{m_1}\hat{n}^d_{m_1\uparrow} - \epsilon_{m_1}\hat{n}^d_{m_1\downarrow} - \epsilon_{m_2}\hat{n}^d_{m_2\uparrow} - \epsilon_{m_2}\hat{n}^d_{m_2\downarrow} \\ &= (\epsilon_{m_1} - \epsilon_{m_2})\left(\hat{n}^d_{m_2\uparrow} + \hat{n}^d_{m_2\downarrow} - \hat{n}^d_{m_1\uparrow} - \hat{n}^d_{m_1\downarrow}\right). \end{aligned} \quad (23)$$

Finally, we substitute these identities into the left-hand side of inequality (12). We have

$$\begin{aligned} &\mathrm{Tr}\left(F(\hat{A}) - F(\hat{B}) - f(\hat{B})(\hat{A} - \hat{B})\right) \\ &= \mathrm{Tr}\Big[- f(\hat{H})\left(\epsilon_{m_1} - \epsilon_{m_2}\right)\Big(\hat{n}^d_{m_2\uparrow} + \hat{n}^d_{m_2\downarrow} \\ &\quad - \hat{n}^d_{m_1\uparrow} - \hat{n}^d_{m_1\downarrow}\Big)\Big] \leq 0. \end{aligned} \quad (24)$$

Dividing both sides of the above inequality by $-\mathrm{Tr}f(\hat{H})$, which is a negative quantity, and using definition (8) for the on-dot single-particle function, we can write the above inequality as

$$(\epsilon_{m_1} - \epsilon_{m_2})\left(n^d_{m_2} - n^d_{m_1}\right) \geq 0. \quad (25)$$

However, according to our starting assumption that $\epsilon_{m_1} > \epsilon_{m_2}$ and $n^d_{m_1} > n^d_{m_2}$ hold simultaneously, the quantity on the left-hand side of the above inequality must be strictly negative. It leads to a contradictory. In other words, the starting assumption must be wrong. It implies that the on-dot single-particle function n^d_m is a non-increasing function of the single-particle energy level index m, as we state in Theorem 1. This completes the proof.

4. Conclusion

In this paper, we study the on-dot single-particle function of a quantum dot system at finite temperature by using a rigorous approach. As an extension of our previous work,[7] we consider the more general case, in which the statistical distribution function is only assumed to be thermodynamically stable. In other words, it is a positive and non-increasing function of the total energy of the quantum dot system. Then, we show that even under this much weaker condition, the non-increasing property of the single-particle function with respect to the on-dot single-particle energy level index m can be re-established too. As a result, our proof gives another example showing that the electronic structure in the quantum dot system is completely determined by its thermodynamic stability. Furthermore, unlike our previous proof, which depends on the explicit expression of the Gibbs distribution function and hence is only applicable to the systems in equilibrium state, the new approach presented in this paper can be potentially applied to the quantum dot systems in non-equilibrium states.

Remark 1 Careful readers may have noticed that the permutation symmetry of the fermion operators in Hamiltonian (1) is essential in establishing Theorem 1. Therefore, a natural question arose is whether this restriction can be removed, since such a symmetry does not generally hold in a real quantum dot system. From the physical point of view, we believe that the single-particle function should be still non-increasing with respect to the on-dot single-particle energy levels, as long as the violation of the permutation symmetry of the fermion operators in the Hamiltonian is not severe. However, so far, we have not found a rigorous approach to prove it. For this purpose, more sophisticated techniques are needed. We shall carry out further investigation on this issue in future.

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