Differences between statistical mechanics and thermodynamics on the mesoscopic scale

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We present a systematic expansion in the ratio between the level spacing and temperature and employ it to evaluate differences between statistical mechanics and thermodynamics in finite disordered systems. These differences are related to spectral correlations in those systems. They are fairly robust and are suppressed at temperatures much higher than the level spacing. [S0163-1829(97)04527-X]

One of the major results that emerged from the work in the field of mesoscopic physics was the understanding that physical quantities may vary *qualitatively* depending on the averaging procedure employed, with a particular emphasis on differences between the canonical (CE) and the grand canonical (GCE) ensembles. This observation became particularly apparent in the study of persistent currents in normal rings[1,2] and has been extended to other thermodynamic[3–5] and transport[6–8] properties. A peculiarity of mesoscopic systems is the existence of an energy E_c associated with spectral correlations on scales larger than the mean level spacing, Δ. Earlier works[9–12] have addressed differences between the ensembles at energies (or temperatures) comparable with Δ. Here we shall study differences between the CE and GCE which persist up to energy scales much larger than Δ. The term ''canonical'' deserves elaboration. One considers an ensemble of finite systems (typically conducting), which are *macroscopically* similar but differ from each other by their impurity configurations, details of the boundaries, etc. We now assign $N^{(i)}$ electrons to the ith member of the ensemble; $N^{(i)}$ is selected arbitrarily, and is uncorrelated with the energy spectrum of the system. Moreover, it is unchanged as we vary some external parameter, x (representing, e.g., an Aharonov-Bohm flux). In particular, one may select $N^{(i)}=N=$ const for all i.

The idea introduced in Ref. 2 was to represent each member of the canonical ensemble as a grand canonical system with an *effective* chemical potential, $\mu^{(i)}(x)$, which is sample specific and depends on the parameter x. A canonically averaged quantity can be obtained by expanding around a ''wrong,'' average, chemical potential, μ (which is independent of i and x), and then calculating the expansion terms (taken at μ, i.e., performed grand canonically). This procedure, leading to remarkable differences between CE and GCE averages, is clearly warranted at zero temperature, $T=0$. In that case differences between the CE and the GCE are solely due to averaging over quenched disorder: each system individually can be equally described by either N or $\mu^{(i)}(x)$, such that[13]

$$-\left.\frac{\partial\Omega^{(i)}}{\partial\mu}\right|_{\mu^{(i)}(x)} \equiv N(\mu^i(x))=N. \tag{1}$$

Here $\Omega^{(i)}(\mu,x)$ is the (sample specific) thermodynamic (TD) potential. The procedure alluded to above has also been employed at $T>0$, defining the sample specific $\mu^i(x)$ through Eq. (1). In that case a canonical system characterized by a fixed number of particles, N, is replaced by a grandcanonical system whose expectation value of the number of electrons is N (at any given value of x). According to this picture differences between the CE and the GCE are again due to quenched disorder (but may depend on temperature). The starting point here is the TD relation

$$F^{(i)}(N,x)=\Omega^{(i)}(\mu,x)+\mu N \tag{2}$$

($F^{(i)}$ is the free energy), leading to the identity

$$\left.\frac{\partial F^{(i)}}{\partial x}\right|_N = \left.\frac{\partial\Omega^{(i)}}{\partial x}\right|_{\mu^{(i)}(x)}, \tag{3}$$

where N is related to $\mu^{(i)}(x)$ through Eq. (1). We should recall, though, that the TD relations, Eqs. (2) and (3), are only approximate [see Eq. (6) below] when it comes to statistical mechanics (SM). It is the latter which should be employed to calculate expectation values of equilibrium observables. $F^{(i)}$ should be derived microscopically, and *not* by calculating $\Omega^{(i)}$ microscopically and then employing Eqs. (1), (2). To understand what is missing in the procedure described above, one may take as an example a system of noninteracting electrons: the occupation probability for a canonical system is *not* given by the Fermi-Dirac function with an effective μ.[5,9,11,12] Differences between canonically and grand canonically averaged quantities should reflect two elements: (i) sample-to-sample fluctuations due to quenched disorder; (ii) the deviation of SM from TD resulting from the fact that the probability of finding the system in a given quantum state differs between the GCE and the CE. The latter element, manifested in corrections to Eqs. (2) and (3), occurs in finite systems[14] and only at finite T.

The purpose of the present work is to elucidate some basic questions concerning the thermodynamics of finite systems. We report on a systematic study of differences between the CE and the GCE, yielding both the contributions due to quenched disorder and due to differences between SM and TD. We specifically consider noninteracting electrons moving in a diffusive disorder at $T>\Delta$. We demonstrate our approach by evaluating both the persistent current and the heat capacity. We indicate how both types of contributions reflect spectral correlations in the system. Our analysis also

points out how the thermodynamic limit (where differences between SM and TD vanish) is approached. Our general scheme should allow for the study of other regimes of disorder (e.g., the dirty ballistic), other types of averaging (e.g., over energy), and the inclusion of electron-electron interactions.

The canonical partition function is given by

$$Z^{(i)}(N) = \mathrm{Tr}\{\delta(\hat{N}-N)e^{-\beta\hat{H}^{(i)}}\} = \frac{1}{2\pi T}\int_{-i\pi T}^{i\pi T} d\mu e^{-\beta[\Omega^{(i)}(\mu)+\mu N]}, \quad (4)$$

where hatted quantities are operators, $e^{-\beta\Omega} \equiv \mathrm{Tr}\{e^{-\beta(\hat{H}-\mu\hat{N})}\}$ and $\beta = 1/k_B T$. Since the eigenvalues of $\hat{N}$ are integers, the δ function is, in fact, a Kronecker function. We note that the evaluation of the integral in Eq. (4) within the saddle point approximation,[15,16] leads to the TD relation Eq. (2). The saddle point $\overline{\mu^{(i)}}(x)$ is sample specific. Instead we expand around a constant $\overline{\mu}$, defined by $\langle N^{(i)}(\overline{\mu})\rangle = N$, where $\langle\ \rangle$ denotes averaging over realizations of the disorder. Defining[17] $\mu - \overline{\mu} \equiv i\sqrt{\Delta T}\tau$ and $V_n^{(i)} \equiv (n!T)^{-1}(-\Delta T)^{n/2}\partial^{n-1}\delta N^{(i)}/\partial\overline{\mu}^{n-1}$ we obtain

$$Z^{(i)}(N) = \sqrt{\frac{\Delta}{2\pi T}} e^{-\beta[\Omega^{(i)}(\overline{\mu})+\overline{\mu}N]} \times \int_{-\pi\sqrt{T/\Delta}}^{\pi\sqrt{T/\Delta}} \frac{d\tau}{\sqrt{2\pi}} e^{-\tau^2/2} \exp\left[\sum_{n=1}^{\infty} V_n^{(i)}\tau^n\right]. \quad (5)$$

Here $\delta N^{(i)}(\overline{\mu},x) \equiv -\partial\Omega^{(i)}/\partial\mu|_{\overline{\mu}} - N$ is the *sample specific* deviation from the mean number of electrons. For $\Delta/T \ll 1$ we may replace the integration limits in Eq. (5) by $\pm\infty$. Expanding the term $\exp[\Sigma_n V_n^{(i)}\tau^n]$, we obtain a diagrammatic expansion where the $\{V_n^{(i)}\}$ play the role of nth order vertices (see below). We stress that the expansion around $\overline{\mu}$ is *not* a standard one: $\overline{\mu}$ does not represent the sample specific saddle point. This expansion is justified *a posteriori* (see below). Employing linked cluster diagrammatic expansion,[18] the free energy may be written

$$F^{(i)}(N) = [\Omega^{(i)}(\overline{\mu}) + \overline{\mu}\overline{N}] + \frac{T}{2}\ln\left(\frac{2\pi T}{\Delta}\right) - \{\text{all connected diagrams}\}^{(i)}. \quad (6)$$

The first term in Eq. (6) yields sample specific grand canonical observables. The second term due to Gaussian fluctuations around the expansion point at $\overline{\mu}$, is a T-dependent sample-independent contribution.

We next consider the diagrammatic expansion of $F^{(i)}(N)$. This expansion involves integrals over τ. In a somewhat artificial, but convenient, analogy with standard perturbation theory, we refer to the term $(2\pi)^{-1/2}\int d\tau \exp\{-\tau^2/2\}\tau^2 = 1$ as a contraction or a ''free propagator,'' symbolically $\langle\tau\tau\rangle$. Similarly a factor $(2\pi)^{-1/2}\int d\tau \exp\{-\tau^2/2\}\tau^4 = 3$ involves three different ways of contraction, $\langle\tau\tau\tau\tau\rangle = 3$. Our ''propagators,'' or *statistical lines*, carry neither energy nor momentum, and are employed to identify contributing diagrams and to evaluate combinational factors. Each propagator is then represented by a ''statistical line'' and each factor V_n corresponds to an nth order vertex, representing an electron loop with n scalar vertices. Examples of connected diagrams contributing to $F^{(i)}(N)$ are shown in Figs. 1, 2. These are skeleton diagrams which are yet to be dressed by impurity lines, interaction lines, etc.

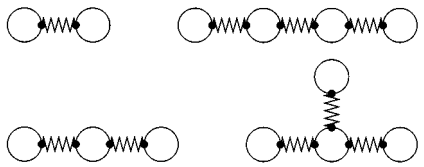

FIG. 1. The $w=0$ family of skeleton canonical diagrams with $k=1,2,3$. The zigzag lines are ''statistical lines;'' full lines — electron propagators; black dots — scalar vertices. The first diagram upon averaging over disorder yields an Altshuler-Shklovskii result.

The following set of rules applies for the calculation of the skeleton diagrams. (1) Consider a connected diagram consisting of p loops with $n_1, \ldots, n_p$ scalar vertices, respectively. We require $\Sigma_{i=1}^{p} n_i = 2k$, with k being the number of statistical lines, $k \geq p-1$. (2) Each statistical line carries a factor $(-\Delta)$. (3) Each n-vertex loop carries a factor $1/n!(\partial^{n-1}/\partial\overline{\mu}^{n-1})\delta N|_{\overline{\mu}}$. (4) Each diagram carries a factor T^w, where $w \equiv k-p+1 \geq 0$. (5) A factor of $1/m!$ should be assigned to any subset of m loops having the same number of vertices. (6) Each diagram should be multiplied by a combinational factor reflecting the number of different ways to interconnect vertices by lines, which result in the very same diagram.

We now divide the skeleton diagrams into families, characterized by the index w. Examples are depicted in Figs. 1, 2. It is understood that disorder averaging has to be carried out subsequently. Let us first consider the $w=0$ family, Fig. 1. It turns out that these are the contributions (describing differences between the CE and the GCE), obtained when Eqs. (2) and (3) are assumed to hold and each member of the canonical ensemble is assigned an effective sample specific chemical potential: the $w=0$ family represents contributions due to quenched disorder, but *not* due to differences between SM and TD. (It is the only contribution which survives at $T=0$.) After some algebra one obtains [hereafter we suppress index (i) and consider only ensemble averaged quantities]

$$\langle\delta F_{w=0}\rangle = -\sum_{k=1}^{\infty} \frac{(-\Delta)^k}{(k+1)!} \frac{\partial^{k-1}}{\partial\overline{\mu}^{k-1}} \langle\delta N^{k+1}\rangle. \quad (7)$$

Exactly the same result may be obtained by the direct solution of Eqs. (1) and (2).[19] The first $(k=1)$ term in the sum

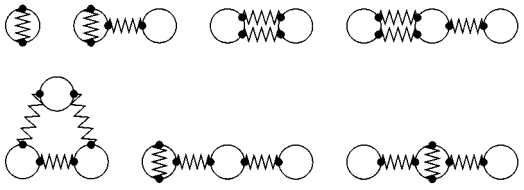

FIG. 2. Skeleton diagrams of the $w=1$ family (up to $k=3$).

corresponds to the two-loop diagram in Fig. 1. Upon averaging over (diffusive) disorder, it yields the Altshuler-Shklovskii term,[20] which has been employed in Ref. 2. The $k\geqslant 2$ terms are given by complete derivatives with respect to $\overline{\mu}$, and are negligible upon averaging (being small in the parameter $\Delta/\overline{\mu}$). This provides us with an *a posteriori* justification of the diagrammatic expansion: sample specific terms in this expansion are not necessarily small, but the ensemble average is well behaved.

We next include the $w\geqslant 1$ families too. These yield contributions due to deviations of SM from TD. It can be shown that for the regime where we employ our expansion, $T>\Delta$, the leading term (in Δ/T) of each family is represented by a two-loop diagrams (cf. Figs. 1 and 2). Evaluation of these diagrams (neglecting full derivatives with respect to $\overline{\mu}$) results in

$$\langle \delta F_{\text{two-loop}}\rangle = \frac{\Delta}{2}\sum_{w=0}^{\infty}\frac{(\Delta T)^w}{(w+1)!}\left\langle\left(\frac{\partial^w \delta N}{\partial \overline{\mu}^w}\right)^2\right\rangle. \tag{8}$$

The $w=0$ term is the Altshuler-Shklovskii quenched disorder contribution;[20] $w=1$ is the leading SM vs TD contribution.[21]

To evaluate these terms we define the correlator K and its Fourier transform $\widetilde{K}$ (see Ref. 22 for a recent review of spectral correlations in disordered systems):

$$\begin{aligned} K(\epsilon-\epsilon') &\equiv \Delta^2[\langle \nu(\epsilon)\nu(\epsilon')\rangle - \langle \nu(\epsilon)\rangle\langle \nu(\epsilon')\rangle] \\ &\equiv \frac{1}{2\pi}\int_{-\infty}^{+\infty} dt \widetilde{K}(t) e^{it(\epsilon-\epsilon')/\Delta}, \end{aligned} \tag{9}$$

where $\nu(\epsilon)$ is the sample specific density of states and t is the dimensionless time (in units of $\hbar/\Delta$). We can write

$$\begin{aligned} \left\langle\left(\frac{\partial^w \delta N}{\partial \overline{\mu}^w}\right)^2\right\rangle = \frac{\partial^w}{\partial \overline{\mu}^w}\frac{\partial^w}{\partial \overline{\mu'}^w}\int\int_{-\infty}^{+\infty}\frac{d\epsilon d\epsilon'}{\Delta^2} f(\epsilon-\overline{\mu}) \\ \times f(\epsilon'-\overline{\mu'})K(\epsilon-\epsilon')|_{\mu'=\mu}, \end{aligned} \tag{10}$$

where f is the Fermi-Dirac function. Changing variables to $\xi\equiv\epsilon-\epsilon'$, $\eta\equiv(\epsilon+\epsilon')/2$, performing the integral over η, and Fourier transforming with respect to ξ, we obtain

$$\langle \delta F_{\text{two-loop}}\rangle = \frac{\pi T}{2}\sum_{w=0}^{\infty}\frac{1}{(w+1)!}\left(\frac{\Delta}{T}\right)^w\int_0^{\infty} dt \frac{t^{2w}}{\sinh^2\pi t}\widetilde{K}\left(t\frac{\Delta}{T}\right). \tag{11}$$

Equations (6) and (11) form the basis for the analysis of the various corrections to the GCE averages, and depict the dependence of these corrections on spectral correlations. We study two examples comparing the $w=0$ (quenched disorder contribution) and $w=1$ (leading contribution due to differences between SM and TD).

The average *persistent current* in the canonical ensemble, $\langle I\rangle_{\text{CE}}$, is obtained by deriving $\langle F\rangle$ with respect to the Aharonov-Bohm flux, ϕ. The flux dependent part of the time correlator for quasi-one-dimensional rings is given by

$$\widetilde{K}(t) = \frac{|t|}{\pi}\sqrt{\frac{1}{4\pi g|t|}}\sum_{p=1}^{\infty} e^{-(p^2/4g|t|)}\cos 4\pi p\phi/\phi_0, \tag{12}$$

where $g=E_c/\Delta\gg 1$ is the dimensionless conductance (E_c is the Thouless correlation energy) and $\phi_0=hc/e$. Only even harmonics appear in Eq. (12). Expanding $\langle I\rangle_{\text{CE}} = \sum_{p=1}^{\infty} I_p \sin 4\pi p\phi/\phi_0$, we find that the quenched disorder contribution is[2]

$$I_p^{\text{dis}} = \frac{\Delta}{\phi_0}\begin{cases} \dfrac{2}{\pi}, & \Delta<T<E_c/p^2 \\ p^2\dfrac{T}{E_c}e^{-\sqrt{(2\pi T/E_c)}p}, & T>E_c/p^2, \end{cases} \tag{13}$$

while the leading SM vs TD correction is given by

$$I_p^{\text{SM-}TD} = \frac{\Delta}{\phi_0}\frac{1}{g}\begin{cases} \dfrac{15\sqrt{2\pi}\zeta(5/2)}{64\pi^3}p\sqrt{\dfrac{E_c}{T}}, & \Delta<T<E_c/p^2 \\ \dfrac{1}{16\pi}p^4\dfrac{T}{E_c}e^{-\sqrt{(2\pi T/E_c)}p}, & T>E_c/p^2. \end{cases} \tag{14}$$

The dependence of the SM-TD contribution on temperature and flux is significantly different from the contribution due to quenched disorder [Eq. (13)], which has been found previously. The former shows that differences between SM and TD decrease slowly (algebraically) with a temperature up to $T\sim E_c\gg\Delta$, and then are suppressed exponentially.

The average canonical *heat capacity* ($\langle C\rangle_{\text{CE}}$) is given by $\langle C\rangle_{\text{CE}} = -\beta^2\partial^2(\beta\langle F\rangle)/\partial\beta^2$ and is written as a sum of contributions [cf. Eq. (6)]:

$$\langle C\rangle_{\text{CE}} = \langle C\rangle_{\text{GCE}} + \langle \delta C^{\text{Gauss}}\rangle + \langle \delta C^{\text{dis}}\rangle + \langle \delta C^{\text{SM-TD}}\rangle. \tag{15}$$

The first term is the grand canonical contribution, which for a degenerate gas of noninteracting electrons is $(\pi^2/3)(T/\Delta)$. The term due to Gaussian fluctuations yields $-\frac{1}{2}$. This contribution can be reinterpreted as a shift of the grand canonical temperature ($T>\Delta$) towards a lower temperature, $T\to T-(3/2\pi^2)\Delta$ forced by the canonical constraints. A similar shift has been found in Ref. 9. The next terms in Eq. (15) are evaluated employing Eq. (11). We are interested in energies larger than Δ, hence $t<1$. The time correlator has two interesting regimes (random matrix theory and Altshuler-Shklovskii,[20] respectively):

$$\widetilde{K}(t) = \frac{|t|}{b\pi}\begin{cases} 1, & g^{-1}<t<1 \\ (4\pi g|t|)^{-d/2}, & \Delta\tau_{\text{el}}<t<g^{-1}, \end{cases} \tag{16}$$

where $b=1,2,4$ for the orthogonal, unitary, and symplectic ensembles, respectively. Here τ_{el} is the elastic mean free time. Equation (16) does not account for the crossover regimes. This leads to

$$\langle \delta C^{\text{dis}}\rangle = \frac{1}{b\pi}\frac{\Delta}{T}\begin{cases} \dfrac{1}{2\pi}, & \Delta<T<E_c \\ \gamma_d\left(\dfrac{T}{E_c}\right)^{d/2}, & T>E_c \end{cases} \tag{17}$$

and in the leading order in Δ/T (Ref. 23),

$$\langle \delta C^{\text{SM-TD}} \rangle = \frac{1}{b\pi}\left(\frac{\Delta}{T}\right)^2 \begin{cases} \dfrac{3\zeta(3)}{4\pi}, & \Delta < T < E_c \\ \eta_d \left(\dfrac{T}{E_c}\right)^{d/2}, & T > E_c . \end{cases} \tag{18}$$

In this example the SM-TD contribution is parametrically smaller than the contribution due to quenched disorder. Here too, differences between SM and TD decay slowly over scales larger than Δ: as a power law, at least over energy up to $\hbar/\tau_{\text{el}}$. The results depicted in Eqs. (17) and (18), as well as an evaluation of $\langle \delta C^{\text{SM-TD}} \rangle$ for a Poissonian spectrum[24] lead us to conclude that the *less* rigid the spectrum the slower the decay with temperature.

In summary, we have presented a systematic expansion in Δ/T describing differences between statistical mechanics and thermodynamics of finite systems. These differences, related to spectral correlations, are found to be fairly robust and slowly suppressed with T. The thermodynamic limit where they are suppressed is attained at temperatures much higher than the level spacing.

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[13] Note that in the grand canonical case, at $T=0$, there are no dynamical fluctuations due to an exchange of particles between the system and the reservoir.

[14] It should be stressed that equilibrium quantities, both within thermodynamics and statistical mechanics, are perfectly well defined even for small systems, as long as the reservoir involved is infinite.

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[21] The two-loop diagram of $w=2$ family [Eq. (8)] is parametrically comparable to the three-loop diagram of $w=1$ family (the latter comes with a small numerical value). Similar statements hold for higher-loop diagrams, which cause deviations of exact result from the one given by Eq. (8). Below we consider only the first two terms of the expansion, which are given accurately by Eq. (8).

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[23] The coefficients are $\gamma_d = \pi^{-1}2^{-(3+d)}d(2-d)\int_0^\infty dx x^{1-d/2}(x^{-2} - \sinh^{-2}x)$, $\eta_d = \pi^{-3}2^{-(6+d/2)}(4-d)(2-d)\Gamma(4-d/2)\zeta(3-d/2)$.

[24] For a Poisson statistics the corrections to C are small (proportional to the inverse bandwidth) but linear in T, in conjunction with this observation.