Persistent Differences between Canonical and Grand Canonical Averages in Mesoscopic Ensembles: Large Paramagnetic Orbital Susceptibilities

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Disorder-averaged thermodynamic quantities within the canonical ensemble are expressed in terms of fluctuations in the grand canonical ensemble, and are then evaluated for the diffusive regime. The particular example of persistent currents in Aharonov-Bohm geometries is addressed, and the harmonics of the disorder-averaged current (which is $\frac{1}{2}\Phi_0$ periodic) are obtained. The orbital susceptibility of a mesoscopic system, disorder averaged over the canonical ensemble, may be large and positive, even for systems which, on a larger scale, should exhibit Landau diamagnetism.

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Following a decade of intense research on the physics of condensed-matter mesoscopic systems[1] one has been led to expect novel phenomena in such systems. These effects usually vanish in the limit when all dimensions of a sample become macroscopic. Among them there are Aharonov-Bohm (AB) oscillations of the conductivity as well as various mesoscopic fluctuation effects. An example of particular interest has been that of the persistent circulating current induced in a mesoscopic ring by an AB flux Φ.[1-9] An experimental demonstration of the possible existence of such currents has been recently reported.[10]

Most mesoscopic effects vanish when averaged over the "*disorder ensemble*" consisting of many macroscopically identical systems, each having its own specific, quenched, defect configuration. In particular cases, however, such effects can have a correlated phase in different members of the disorder ensemble. In such situations, the signals associated with these ensemble members will add up coherently rather than average to a vanishing magnitude per sample. A notable example for such a survival has been the $(h/2e)$-periodic conductance oscillation as a function of the AB flux in mesoscopic cylinders or arrays of rings.[11,12] This suggests a fundamental question of what are the conditions under which a given mesoscopic effect will survive the disorder-ensemble averaging. In the particular case of the $h/2e$ oscillations of the conductance the coherent backscattering is the relevant mechanism.[11] It is this survival which permitted the observation of these oscillations in, e.g., macroscopically long cylinders to be made.[12]

In the case of persistent currents induced by an AB flux, only an ensemble-averaged effect could be observed in the experiments of Ref. 10 which dealt simultaneously with 10^7 mesoscopic rings. Of particular fundamental interest is the observation[9] that in this case the $h/2e$ harmonic will survive but *only* in the canonical ensemble (i.e., when the number of electrons in *each member* of the disorder ensemble is kept fixed and flux independent) in contrast with the grand canonical ensemble (in which the chemical potential is fixed in *each sample* while the number of electrons is allowed to vary with the flux). This observation was first made in the 1D case in Ref. 6. Bouchiat and Montambaux[9] derived it for finite-cross-section rings with very weak and very strong disorder. They also observed the difference between canonical and grand canonical ensembles in numerical simulations at intermediate disorder and realized its generality.

This paper is devoted to the above question, using the AB persistent current as an example, but the idea is much more general. We derive an expression for the difference between canonical and grand canonical disorder-ensemble averages. The latter averages are known[7,8] to be exponentially small for the problem at hand. Using this expression we have *determined* the average Fourier components of the persistent current $I(\Phi)$, which imply some nontrivial properties of the electronic energy spectra as a function of Φ.

While the results we obtained come short by almost 2 orders of magnitude from explaining the experimental results of Ref. 10, they lead to new and unexpected conclusions for, e.g., the orbital susceptibility of a macroscopic 2D electron gas when it is divided into many small parts, of size scale L each. We now consider the diffusive case. Provided the number of electrons in each of the parts is *kept constant,* the orbital susceptibility χ should depend on L as follows: Once L becomes comparable to the phase coherence length L_ϕ (i.e., $\hbar/\tau_\Phi=\hbar D/L_\Phi^2$ is comparable to the Thouless energy $E_c=\hbar D/L^2$, where D is the electron diffusion constant), χ *changes sign* and remains of the order of the Landau susceptibility χ_L in 2D. With further decrease of L, χ increases to values of order $\chi_L E_c/\Delta \gg \chi_L$ for $\Delta \sim \hbar/\tau_\Phi$, Δ being the mean level spacing at the Fermi energy. At finite temperature T, $\hbar/\tau_\Phi$ should be replaced by $\max\{T,\hbar/\tau_\Phi\}$. It is straightforward to generalize the above to finite-thickness slabs.

We start by deriving expressions for various averages of the free-energy derivatives with respect to the AB flux Φ. Let $F(N,\Phi)$ be the free energy in the canonical ensemble and $\Omega(\mu,\Phi)$ be the grand potential (N is the number of electrons and μ is the chemical potential).[13] From thermodynamics,

$$\left(\frac{\partial F}{\partial \Phi}\right)_N = \left(\frac{\partial \Omega}{\partial \Phi}\right)_{\mu=\partial F/\partial N|_\Phi} = -I(\Phi)\,, \tag{1}$$

where we take $c=1$. A crucial point for our analysis is that in the canonical ensemble μ can depend on Φ, and vary from sample to sample,

$$\mu = \langle\mu\rangle + \delta\mu(\Phi)\,, \tag{2}$$

where $\langle\,\rangle$ stands for the disorder-ensemble average. From (1) we have

$$\left(\frac{\partial F}{\partial \Phi}\right)_N = \left(\frac{\partial \Omega}{\partial \Phi}\right)_{\mu=\langle\mu\rangle} + \delta\mu\left[\frac{\partial}{\partial\mu}\left(\frac{\partial \Omega}{\partial \Phi}\right)_\mu\right]_{\mu=\langle\mu\rangle} \tag{3}$$

provided $\delta\mu$ is small enough. Now we change the order of derivatives and average over the ensemble. As a result, since $\langle(\partial\Omega/\partial\Phi)_\mu\rangle \approx 0$,

$$\left\langle\left(\frac{\partial F}{\partial \Phi}\right)_N\right\rangle = \left\langle\delta\mu\frac{\partial}{\partial\Phi}\left(\frac{\partial \Omega}{\partial \mu}\right)_\Phi\right\rangle = -\left\langle\delta\mu\left(\frac{\partial N}{\partial \Phi}\right)_N\right\rangle_{\mu=\langle\mu\rangle}. \tag{4}$$

We have used the identity $N=-\partial\Omega/\partial\mu$. $\delta\mu$ can be easily connected with δN—the number-of-particle variance in the corresponding grand canonical ensemble with $\mu=\langle\mu\rangle$, $\delta\mu=\delta N(\partial N/\partial\mu)_\Phi^{-1}$. Thus,

$$\left\langle\left(\frac{\partial F}{\partial \Phi}\right)_N\right\rangle = -\left\langle\left(\frac{\partial N}{\partial \mu}\right)_\Phi^{-1}\delta N\frac{\partial N}{\partial\Phi}\right\rangle_{\mu=\langle\mu\rangle}, \tag{5}$$

where averaging on the right-hand side is taken at fixed $\mu=\langle\mu\rangle$. Since $N(\mu=\langle\mu\rangle)$ is our fixed, Φ-independent N, we can substitute $\partial(\delta N)/\partial\Phi$ for $\partial N/\partial\Phi$:

$$\left\langle\left(\frac{\partial F}{\partial \Phi}\right)_N\right\rangle = -\frac{1}{2}\left\langle\left(\frac{\partial N}{\partial \mu}\right)^{-1}\frac{\partial(\delta N)^2}{\partial\Phi}\right\rangle_{\mu=\langle\mu\rangle} = -\frac{\Delta}{2}\frac{\partial}{\partial\Phi}\langle(\delta N)^2\rangle_{\mu=\langle\mu\rangle}\,. \tag{6}$$

For the last equality we used $\langle(\partial N/\partial\mu)^{-1}\rangle=\Delta$, which is valid up to higher-order corrections in $g=E_c/\Delta$.

The right-hand side of (6) can also be written as $(2\Delta)^{-1}\partial\langle(\delta\mu)_N^2\rangle/\partial\Phi$; this form had been less generally derived in Ref. 14. The variation of μ with Φ in a given sample at $T\to 0$ is similar to that of a typical level. Thus the averaged total current is proportional to the mean-squared fluctuation of a single-level current. This prediction for the lowest harmonic of $I(\Phi)$ agrees with a strong disorder result of Ref. 9. The general validity of these results was also checked numerically.[9]

We have derived Eq. (6) for the particular case of persistent current in a ring with AB flux Φ. In fact, both Eq. (3) and the equality of its last term with the middle expression of Eq. (6) are completely general. They hold for appropriate derivatives of the free energies with respect to any external parameter for arbitrary sample geometry and including all interactions.[15,16] The usefulness of Eq. (6) is in enabling us to evaluate canonical averages using grand canonical ones which are usually much easier to compute.

We now apply Eq. (8) to calculate $I(\Phi)$ in the diffusive regime, using also the results of Ref. 17 for $\langle(\delta N)^2\rangle$,

$$\langle(\delta N)^2\rangle = \int\int_0^\mu d\epsilon_1\, d\epsilon_2 K(\epsilon_1,\epsilon_1)\,, \tag{7}$$

where the correlation function of the densities of states $K(\epsilon_1,\epsilon_2)$ at small enough $\epsilon_-=\epsilon_1-\epsilon_2$ (i.e., $\epsilon_1-\epsilon_2 \ll 1/\tau$, where τ is the time of a mean free path, $\mu\tau\ll 1$, $\hbar=1$) equals

$$K(\epsilon_1,\epsilon_2) = -\frac{s^2}{\pi^2}\mathrm{Re}\sum_{n_\alpha}\left(\epsilon_- + \frac{i}{\tau_\Phi} + iDq^2\right)^{-2}. \tag{8}$$

Here α can be x, y, or z; s is the degeneracy (say, spin), and for a sample with dimensions $L_x\times L_y\times L_z$ (L_x is a ring perimeter), $q^2=\pi^2\sum_{\alpha=x,y,z}(n_\alpha/L_\alpha)^2$. At zero flux $n_\alpha=0, \pm1, \pm2, \ldots$. Equation (8) is the result of the summation of the usual diffusion and Cooperon diagrams.[17] We can consider the correlation of the number of particles at different Φ (Φ_1 and Φ_2). In this case we can keep using Eq. (8) but for the diffusion contribution, $n_x=\text{integer}+(\Phi_1-\Phi_2)/\Phi_0$, while for Cooperon contribution, $n_x=\text{integer}+(\Phi_1+\Phi_2)/\Phi_0$, where $\Phi_0=\hbar/e$.

For the calculation of the derivative (8) we consider the difference $\langle[\delta N(\Phi)]^2-[\delta N(0)]^2\rangle$. For this difference $K(\epsilon_1,\epsilon_2)$ can be taken in the form (8) since this difference is determined by ϵ_1,ϵ_2 close to the Fermi level. Since $\Phi_1=\Phi_2$, the flux dependence of the diffusion contributions cancel while the Cooperon contributions lead to

$$\frac{\partial}{\partial\Phi}\langle[\delta N(\Phi)]\rangle^2 = -\mathrm{Re}\frac{\partial}{\partial\Phi}\sum_{n_\alpha}\frac{s^2}{2\pi^2}\int\frac{d\epsilon_1 d\epsilon_2}{\{\epsilon_- + i/\tau_\Phi + iD\pi^2[n_y^2/L_y^2 + n_z^2/L_z^2 + (n_x+2\Phi/\Phi_0)^2/L_x^2]\}^{-2}}\,. \tag{9}$$

It is clear from (9) that $\langle(\partial F/\partial\Phi)_N\rangle$ is periodic in Φ with a period $\Phi_0/2$:

$$\left\langle\left(\frac{\partial F}{\partial \Phi}\right)\right\rangle = -\sum_{m=-\infty}^{\infty} I_m \exp\left(i4\pi\frac{m\Phi}{\Phi_0}\right). \tag{10}$$

For simplicity, let us consider the 1D case where $L_y,L_z\to 0$ so that DL_y^{-2} is much larger than $E_c=DL_x^{-2}$. Then in the sum (8) all terms with nonzero n_y or n_z can be neglected. Performing the sum in Eq. (8) with the Poisson sum formula we find for I_m,

$$I_m = \frac{s^2\Delta}{4\pi^3} \int\int d\epsilon_1 d\epsilon_2 \int_{-\infty}^{\infty} d\Phi \left[\frac{\partial}{\partial\Phi} \mathrm{Re} \left(\epsilon_- + \frac{i}{\tau_\Phi} + i4\pi^2 E_c \frac{\Phi^2}{\Phi_0^2} \right)^{-2} \right] e^{-4i\pi m\Phi/\Phi_0} . \tag{11}$$

We change the integration variables to $\epsilon_\pm = \epsilon_1 \pm \epsilon_2$ and find

$$I_m = \frac{-ims^2\Delta}{4\pi^2 E_c} \int_{-\infty}^{0} d\epsilon_+ \frac{d}{d\epsilon_+} \frac{E_c}{|m|} \left\{ \exp\left[-2im \left(\frac{i\epsilon_+ \tau_\Phi - 1}{E_c \tau_\Phi} \right)^{1/2} \right] + \exp\left[-2im \left(\frac{-i\epsilon_+ \tau_\Phi - 1}{E_c \tau_\Phi} \right)^{1/2} \right] \right\} , \tag{12}$$

where the imaginary parts of the roots should be negative at $m > 0$ and positive at $m < 0$. As a result

$$I_m = \frac{is^2\Delta}{\pi\Phi_0} \exp\left(\frac{-2m}{(E_c \tau_\phi)^{1/2}} \right) \mathrm{sgn}\, m . \tag{13}$$

Therefore, for $E_c \tau_\Phi > m^2$, I_m is *independent of* E_c (i.e., of *disorder*) *and of m,* and *it is of order* Δ. Near $\Phi = 0$

$$\left\langle \left(\frac{\partial^2 F}{\partial \Phi^2} \right)_N \right\rangle_{\Phi=0} = \frac{8\pi^2 i}{\Phi_0^2} \sum_{m=-\infty}^{\infty} m I_m = 2s^2 \Delta \frac{E_c \tau_\Phi}{\Phi_0^2} . \tag{14}$$

This is valid for $E_c \tau_\Phi \gg 1 \gg \Delta \tau_\Phi$. Equation (14) can also be obtained directly from Eq. (9). We note that when τ_ϕ^{-1} decreases from $O(E_c)$ to $O(\Delta)$ the curvature (14) increases from $O(\Delta)$ to $O(E_c)$. The latter value is the expected order of magnitude[18,19] as $\tau_\Phi \to \infty$.

To summarize our results: A $(h/2e)$-periodic paramagnetic persistent current is obtained in the canonical disorder-ensemble average. Its $(E_c/\Delta)^{1/2}$ first harmonics are equal to $4e\Delta/\pi^2\hbar$ for $T, \tau_\Phi \lesssim \Delta$. With increasing τ_Φ^{-1} and/or T the harmonics are progressively cut off with only the lowest ones remaining at $T, \tau_\Phi^{-1} \sim E_c$. The maximal amplitude of the average current decreases from $O(e(E_c\Delta)^{1/2}/\hbar)$ to $O(e\Delta/h)$ when T and τ_Φ^{-1} increase from $\lesssim \Delta$ to E_c. The variations of $\delta\mu$ and a typical single-level energy with Φ are of $O(\Delta)$. Since the phases of the harmonics $\delta\mu(\Phi)$ are random, the random fluctuations of $\delta\mu$ are over a flux scale of $O(\Phi_0(\Delta/E_c)^{1/2})$. This leads to a typical single-level current[8] of $O(e(E_c\Delta)^{1/2}/\hbar)$. It is interesting to note that the flux-independent part of the fluctuation $\langle \delta N^2 \rangle$ found in Ref. 17 is much larger than the flux-dependent part, $\langle \delta N(\Phi)^2 \rangle_\mu \sim 1$, found here. This implies that the usual "ergodic" hypothesis is not valid for the ensemble versus the flux-dependent fluctuations of the levels.

The size of the persistent currents found here comes short by about 2 orders of magnitude from explaining the experimental results of Ref. 10. Likewise, when appropriate values,[20] of the order of 0.04, are used for the electron-electron interaction constant, the theory based on the latter[15,16] also yields results an order of magnitude smaller than the results of Ref. 10. To explain the latter, several harmonics of $\langle I \rangle(\Phi)$ will have to be assumed, or further mechanisms such as the operation of the charging energy[21,22] or certain properties of the spectrum $\{E_j(\Phi)\}$ (Ref. 22) will have to be invoked. Results very similar to ours were obtained independently by Schmid,[23] who in addition emphasized the role of electron-electron interactions in ironing out changes of the electrons' charge density.

These results are generalizable to the case of singly connected quantum dots. The behavior mentioned at the beginning of this paper can be obtained for the canonical disorder-ensemble average of the orbital magnetic response. Equation (3) and subsequently Eq. (6) hold for a singly connected quantum dot. Employing these relations, one can show (details will be given elsewhere) that the canonical free energy of a quantum dot depends on the flux in the same way as in a ring. For small fluxes this leads to the linear susceptibility described above. This is a novel *mesoscopic effect.* The transition from the bulk diamagnetic response to the atomic one with decreasing L is not monotonic: χ changes sign and increases markedly in an appropriate regime. All of the above is valid when the electrons are *diffusive.* The crossover of χ to possibly large negative values at sizes so small that disorder is irrelevant will be considered elsewhere.

The differences between the canonical and grand canonical behavior found here are due to energy levels crossing a given chemical potential μ from above or from below and their contribution to the canonical energy subtracting from and adding to, respectively, the grand potential. Since the curvatures of such levels having crossed μ from above or below are opposite, they give contributions of the same sign, which is easily seen to be paramagnetic, to χ.

The effect can be understood since Φ breaks time-reversal symmetry and changes the Wigner-Dyson level correlations from orthogonal to unitary. This means, for example, that small energy separations will typically increase (quadratically at $\Phi = 0$) with Φ. It is the *lower* member of each such pair, having a larger thermal population, which has a negative curvature at $\Phi = 0$ as a function of Φ. This[24] causes the average *orbital* susceptibility to be paramagnetic, in marked distinction to the diamagnetic sign found in the ideal, cylindrically symmetric and integrable case.

The spin-orbit interaction is *not* expected to change the sign of the average susceptibility even in the strong spin-orbit scattering limit. This statement does not apply for a strictly one-dimensional system in accordance with the analysis of Ref. 25. This issue as well as the crossover to ideal systems and the role of electron-electron interactions[15,16,21-23] will be discussed elsewhere.

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