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Magnetic susceptibility of ballistic microstructures

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The orbital magnetic susceptibility of ballistic microstructures is considered within the independent-electron model. Using semiclassical theory, specifically Gutzwiller's trace formula, the finite-size corrections to the Landau susceptibility are expressed in terms of the classical periodic orbits. It is found that these finite-size corrections can be much larger than the bulk susceptibility in the quantum-coherent regime. It is demonstrated that the orbital susceptibility is a sensitive probe of quantum chaos, having a larger amplitude in integrable than in completely chaotic ballistic microstructures. The approach is applied to the square billiard. While the predictions for the amplitude and the magnetic-field dependence are consistent with recent experimental results by Lévy *et al.*, the theory predicts a faster decrease with temperature than observed experimentally.

I. INTRODUCTION

In a recent experiment Lévy *et al.*[1] measured the orbital magnetic susceptibility of an ensemble of 10^5 isolated GaAs squares at subkelvin temperatures. The susceptibility was found to exhibit a paramagnetic zero-field peak whose height exceeds the Landau susceptibility by two orders of magnitude, and whose width corresponds to roughly one flux quantum threading each square. With temperature the peak decreases on a scale of about 0.5 K. Motivated in part by this experiment, this paper is concerned with quantum-coherence corrections to the Landau susceptibility within the independent-electron model.

Theoretical work on finite-size corrections to Landau diamagnetism has a long history.[2] Landau[3] showed that the bulk orbital susceptibility of free electrons is independent of magnetic field for temperatures large compared to the spacing of Landau levels. For lower temperatures (or stronger magnetic fields) the susceptibility exhibits de Haas–van Alphen oscillations. Subsequently, Hund[4] argued qualitatively that in finite-size systems there are oscillatory contributions to the susceptibility even for weak magnetic fields. These oscillatory contributions were calculated by Dingle[5] for a cylinder threaded by an axial magnetic field. Treating the magnetic field perturbatively, he found that the spectrum of the susceptibility oscillations is determined by the flux threading arbitrary regular polygons inscribed into the circular cross section of the cylinder. Motivated by experiments on clusters,[6] similar (numerical) perturbative calculations for simple geometries were performed by van Ruitenbeek and van Leeuwen.[7] Smooth, nonoscillatory finite-size corrections to the Landau susceptibility were discussed by Robnik.[8] A number of authors[9–11] considered the orbital magnetic response of mesoscopic samples in the diffusive regime.

Recently, it was shown both theoretically[12] and experimentally[13] that the conductance of ballistic microstructures is sensitive to whether the classical dynamics is integrable or chaotic. It is an interesting question whether this is also true for the susceptibility. This question was first addressed by Nakamura and Thomas,[14] who calculated the susceptibility fluctuations numerically for a family of billiard systems, and argued that they are larger in chaotic than in integrable billiards. Their conclusions were criticized by Németh,[15] who found large oscillations in a simple, integrable model.

My purpose in this paper is to explain how semiclassical methods can be used to calculate oscillatory contributions to the susceptibility analytically for a large class of ballistic microstructures.[16,17] The results generalize those obtained by Dingle[5] for the cylinder geometry. In particular, the approach is first applied to calculate the mesoscopic susceptibility for the ideal square billiard, and the results are compared with experiment. Second, the controversy concerning the amplitude of the susceptibility fluctuations in chaotic and integrable billiard systems is resolved.

The susceptibility is defined in terms of the grand-canonical thermodynamic potential Ω as

$$\chi=-\frac{\partial^2\Omega}{\partial B^2}\ , \tag{1}$$

where B denotes the magnetic field. At zero temperature and within the independent-electron picture, Ω can be expressed in terms of the spectral density $\rho(E)=\Sigma_n\delta(E-e_n)$ (the e_n denoting the single-particle eigenenergies) as

$$\Omega(\mu)=\int_0^\mu dE(E-\mu)\rho(E)\ . \tag{2}$$

Semiclassically, the smooth, energy-averaged spectral density $\langle\rho(E)\rangle=1/\Delta$ is determined by the classical phase-space volume (one state per Planck cell). In mesoscopic systems the discrete nature of the spectrum becomes important, and hence deviations $\delta\rho(E)$ from the average spectral density must be considered. General semiclassical formulas, which express $\delta\rho(E)$ in terms of classical periodic orbits, were derived by Gutzwiller[18] for

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chaotic and by Berry and Tabor[19] for integrable systems. The resulting periodic-orbit contributions to the susceptibility $\delta\chi$ oscillate as a function of magnetic field and chemical potential. Reflecting the quantum-interference nature, the contribution of a particular periodic orbit is significant only if the orbit length is shorter than the phase-coherence length L_Φ and the thermal length L_T. In the following, the periodic-orbit contributions are also referred to as mesoscopic susceptibility.

The semiclassical approach becomes particularly simple for classically weak magnetic fields, for which the cyclotron radius R_c is much larger than the phase-coherence length L_Φ. In this limit the magnetic field does not affect the geometry of the periodic orbits which contribute to the mesoscopic susceptibility. Instead, it enters the thermodynamic potential only through the Aharonov-Bohm phase proportional to the area enclosed by the orbit. For weak magnetic fields the periodic-orbit contributions to the susceptibility are entirely analogous to mesoscopic persistent currents,[4,20] except that the susceptibility is not perfectly periodic with flux because different periodic orbits enclose different areas. A corresponding semiclassical theory of persistent currents was recently developed.[21,22]

It is sometimes assumed that the important energy scale for quantum-size effects on the magnetization is the level spacing.[7] The semiclassical approach shows that, instead, the relevant energies are the orbit correlation energies h/T_γ (T_γ is the traversal time of the orbit), which are usually much larger than the level spacing. In particular, the orbit correlation energies determine the dependences on the chemical potential, inelastic scattering, and temperature.

The semiclassical approach allows one to draw general conclusions about the mesoscopic susceptibility in integrable and completely chaotic billiard systems. For our purposes the main difference between integrable and chaotic systems lies in the nature of the classical periodic orbits. Whereas they form continuous families in the integrable systems, almost all orbits are isolated and unstable in chaotic systems. In particular, I refer to systems as completely chaotic if *all* periodic orbits are isolated and unstable. The contribution to the susceptibility of continuous families of periodic trajectories is enhanced by constructive interference, and hence the mesoscopic susceptibility of integrable microstructures is larger than that of completely chaotic ones. This result contradicts the conclusions of Nakamura and Thomas.[14] Universal amplitudes are derived for integrable and completely chaotic billiards which determine the mesoscopic susceptibility up to (nonuniversal) functions of ratios of geometric length scales. Analogous results were previously obtained for the persistent-current amplitude.[22] Experimentally, the susceptibility is more easily accessible.

Interestingly, I find that the mesoscopic susceptibility typically increases faster with system size than the bulk Landau susceptibility. Even though this result may seem puzzling at first because the mesoscopic susceptibility is derived from corrections to the bulk thermodynamic potential, it can be understood as a direct consequence of phase coherence. The Aharonov-Bohm phase couples the magnetic field to the area of the sample, leading to additional "volume" factors in the susceptibility. One may ask whether there are also significant finite-size corrections to the Pauli spin susceptibility.[23] These can be computed within the same semiclassical approach, and one finds that they are much smaller than the orbital susceptibility (Appendix A).

The mesoscopic susceptibility is sample specific, reflecting the details of the periodic orbits of the system, and changes sign as function of magnetic field and chemical potential. This leads to ensemble-averaging questions which are analogous to those for the mesoscopic persistent current.[24] In the ballistic regime one considers an ensemble of systems with different chemical potentials and geometric parameters. The typical mesoscopic susceptibility measured in an experiment on a single sample (e.g., a single square) is characterized by the rms average $\langle \delta\chi^2 \rangle^{1/2}$. The experiment by Lévy *et al.*[1] was performed on an ensemble of 10^5 squares. Hence it requires one to calculate the ensemble-averaged mesoscopic susceptibility $\langle \delta\chi \rangle$. As for the persistent current, the mesoscopic susceptibility is vanishingly small when averaged in the grand-canonical ensemble (fixed chemical potential μ).[25] However, the average is nonzero and paramagnetic for small values of B, if it is performed in the canonical ensemble (fixed number of electrons N).[10,26,27]

Motivated by the experiment of Ref. 1, I calculate the mesoscopic susceptibility of the ideal square billiard. This simple model of the experimental sample neglects deviations from the ideal geometry, scattering due to boundary roughness and residual disorder, which affect the geometry of the periodic orbits, and electron-electron interactions, which may lead to an additional contribution to the susceptibility in analogy to the average persistent current in the diffusive regime.[28] The theoretical predictions for the amplitude and the magnetic-field dependence are consistent with the experimental results of Ref. 1. However, the theoretical result for the susceptibility decreases much faster with temperature than the experimental one.

This paper is organized as follows. The semiclassical approach is developed in Sec. II A. The susceptibility is related to classical periodic orbits, and it is shown that the approach becomes particularly simple in the weak-field limit, where the magnetic field enters predominantly through the Aharonov-Bohm phases. The approach is applied to the square billiard in Sec. II B. The periodic orbits of the square billiard and their contributions to the spectral density are discussed in Sec. II B 1. The typical mesoscopic susceptibility is computed in Sec. II B 2. The ensemble-averaged susceptibility relevant to the experiment by Lévy *et al.* is calculated and discussed in Sec. II B 3. The predictions are compared with the experiment in Sec. II B 4. General intregrable and chaotic billiards are considered in Sec. II C. The results are summarized and discussed in Sec. III. Various details are considered in three appendices. Appendix A deals with finite-size corrections to the spin susceptibility. The semiclassical spectral density of the square billiard in the absence of a magnetic field is computed in Appendix B.

Finally, the formula for the average mesoscopic susceptibility in the canonical regime is justified carefully in Appendix C.

II. SEMICLASSICAL APPROACH

A. General formulation

In this section the mesoscopic susceptibility is related to the classical periodic orbits. It is shown that the semiclassical approach becomes particularly simple in the weak-field limit, where the magnetic field enters the periodic-orbit contributions to the thermodynamic potential Ω only through the Aharonov-Bohm phase. First the thermodynamic potential is considered in the absence of the magnetic field. The leading contribution to Ω derives from the energy-averaged spectral density $\langle\rho(E)\rangle$. In mesoscopic systems the discrete nature of the spectrum leads to quantum-coherence corrections $\delta\Omega$ to the thermodynamic limit. At finite temperature one has within the independent-electron model

$$\delta\Omega=-\frac{1}{\beta}\int_0^\infty dE\,\delta\rho(E)\ln\{1+\exp[-\beta(E-\mu)]\}$$
$$=-\int_0^\infty dE\,\delta N(E)f_\mu(E)\ , \tag{3}$$

where the deviations from the smooth spectral density are defined as $\delta\rho(E)=\rho(E)-\langle\rho(E)\rangle$. Furthermore, $\beta=1/k_BT$ denotes the inverse temperature, $f_\mu(E)$ is the Fermi function, and $\delta N(E)=\int_0^E dE'\delta\rho(E')$. It is a central result of semiclassical theory that $\delta\rho(E)$ can be expressed as a sum over classical periodic orbits γ (Gutzwiller's trace formula):[18,19]

$$\delta\rho(E)=\frac{1}{\hbar^\nu}\sum_\gamma A_\gamma\exp\left\{\frac{i}{\hbar}S_\gamma(E)\right\}\ . \tag{4}$$

Here $S_\gamma(E)$ denotes the classical action of the periodic orbit. For billiards it is given in terms of the length L_γ of the orbit, $S_\gamma=\hbar kL_\gamma$. The exponent ν and the amplitude A_γ depend on the type of periodic orbit. While $\nu=(d+1)/2$ for the nonisolated periodic orbits of integrable systems (d denotes the dimensionality of the system), one has $\nu=1$ for the unstable and isolated periodic orbits of chaotic systems. Useful expressions for the amplitude A_γ were derived by Gutzwiller[18] for chaotic and by Berry and Tabor[19] for integrable systems. From Eq. (4) one finds

$$\delta N(E)=\frac{1}{i\hbar^{\nu-1}}\sum_\gamma\frac{A_\gamma}{T_\gamma}\exp\{ikL_\gamma\}\ . \tag{5}$$

Inserting this expression into Eq. (3) and linearizing the action around the chemical potential μ, one has

$$\delta\Omega=-\frac{1}{i\hbar^{\nu-1}}\sum_\gamma\frac{A_\gamma}{T_\gamma}\exp\{ik_FL_\gamma\}$$
$$\times\int_0^\infty dE\,\exp\left\{i\frac{T_\gamma}{\hbar}(E-\mu)\right\}f_\mu(E)\ , \tag{6}$$

where

$$T_\gamma=\frac{dS_\gamma}{dE} \tag{7}$$

denotes the orbit traversal time. The energy integral in Eq. (6) is evaluated by contour integration. The Fermi function has poles at $E=\mu+i(2n+1)\pi/\beta$ with residues $-1/\beta$. Since only energies close to the chemical potential are relevant, one can extend the lower limit of integration to $-\infty$. Hence one finds

$$\delta\Omega=\frac{1}{\hbar^{\nu-2}}\sum_\gamma\frac{A_\gamma}{T_\gamma^2}\exp\{ik_FL_\gamma\}\left[\frac{\pi T_\gamma}{\hbar\beta}\right]\sinh^{-1}\left[\frac{\pi T_\gamma}{\hbar\beta}\right]\ . \tag{8}$$

The contribution of each periodic orbit oscillates with chemical potential with a period equal to the orbit correlation energy $h/T_\gamma=hv_F/L_\gamma$. In general, there are infinitely many classical periodic orbits which contribute to Eq. (8). Its usefulness derives from the fact that finite temperature and inelastic scattering rapidly restrict the number of orbits that must be considered. A periodic orbit contributes significantly only if the orbit length L_γ is of the order of or shorter than both the thermal length $L_T=\hbar v_F/k_BT$ and the phase-coherence length L_Φ.

Next, I discuss the effect of the magnetic field. It affects the periodic-orbit contributions to the susceptibility in two ways. The difference in the corresponding magnetic-field scales leads to an important simplification in the weak-field limit. Between reflections from the boundaries of the ballistic microstructures, the (classical) electronic trajectories in a magnetic field are circular arcs of cyclotron radius $R_c=mv/eB$. The corresponding *classical* field scale can be defined by $L_\gamma\simeq R_c$, or, equivalently, in terms of the flux ϕ threading the billiard, $\phi\approx(k_FL/2\pi)(L/L_\gamma)\phi_0$ (here ϕ_0 denotes the flux quantum and L a typical linear dimension of the billiard). The magnetic field also enters through the Aharonov-Bohm phase proportional to the magnetic flux enclosed by the periodic orbit. The corresponding *quantum* scale is much smaller than the classical one, roughly one flux quantum threading the sample. Hence, in the limit $R_c\gg\min\{L_\Phi,L_T\}$ one may neglect the bending of the classical periodic orbits by the magnetic field and retain only the magnetic-field dependence of the Aharonov-Bohm phases.

B. The square billiard

1. Periodic orbits

Motivated by the experiment by Lévy *et al.*[1] I illustrate the general approach outlined in Sec. II A by calculating the mesoscopic susceptibility of the square billiard in the weak-field limit. The primitive periodic orbits (single traversals) of the square of side L can be labeled by two (coprime) integers $\sigma=(m_1,m_2)$ corresponding to the winding numbers of the orbit parallel to the sides of the square. This is shown in Fig. 1. The number of retracings is labeled by p. The lengths of the primitive orbits are

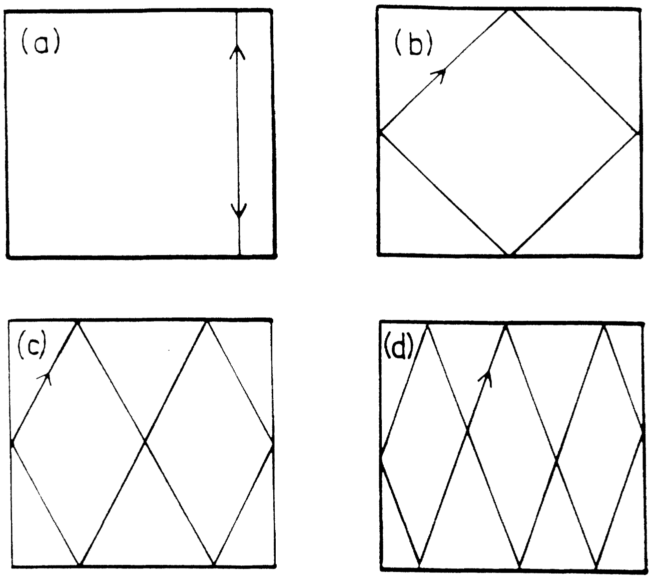

FIG. 1. Primitive periodic orbits of the square billiard, labeled by two (coprime) integers $\sigma=(m_1,m_2)$ corresponding to the winding numbers in the vertical and horizontal directions: (a) (1,0), (b) (1,1), (c) (2,1), and (d) (3,1). Only orbits with both m_1 and m_2 odd enclose area and contribute to the susceptibility.

$$L_\sigma=2L\sqrt{m_1^2+m_2^2}\ . \tag{9}$$

For the square geometry different periodic trajectories belonging to a nonisolated periodic orbit enclose different areas. One finds that primitive orbits with either m_1 or m_2 even enclose zero area and therefore do not contribute to the susceptibility. For orbits with both m_1 and m_2 odd, the periodic trajectories are labeled by their distance x from the self-retracing trajectory as illustrated in Fig. 2. For the areas enclosed by the trajectories one finds by elementary geometry that

$$A_\sigma(x)=4\mathcal{A}_\sigma\frac{x}{x_\sigma}\left[1-\frac{x}{x_\sigma}\right]\ . \tag{10}$$

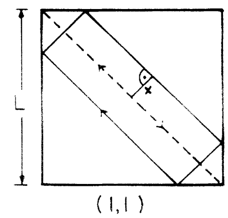

FIG. 2. Areas enclosed by the periodic trajectories contained in the (1,1) orbit of the square billiard. The parameter x denotes the "distance" from the self-retracing orbit (dashed line). The analogous construction for more complicated orbits is tedious but straightforward.

Here the maximum enclosed area is $\mathcal{A}_\sigma=L^2/2m_1m_2$. The area vanishes for $x=0$ and $x_\sigma=L/(m_1^2+m_2^2)^{1/2}$. For retracings of primitive orbits the enclosed area is multiplied by a factor p.

In the absence of the magnetic field the amplitudes of the contributions to the spectral density due to these periodic orbits can be obtained from the general expression due to Berry and Tabor.[19] However, for this simple geometry they can also be found by direct computation based on the exact spectrum. I find

$$\delta\rho(E)=\frac{2^{5/2}}{\pi^{1/2}}\frac{1}{\Delta}\sum_{p=1}^{\infty}p^{-1/2}\sum_\sigma \delta_\sigma\frac{\cos(pkL_\sigma-\pi/4)}{(kL_\sigma)^{1/2}}\ , \tag{11}$$

where Δ denotes the average level spacing, and the factor δ_σ stands for

$$\delta_\sigma=\begin{cases}\frac{1}{2} & \text{if } m_1=0 \text{ or } m_2=0\\ 1 & \text{otherwise}\ .\end{cases} \tag{12}$$

Details of the calculation are shown in Appendix B.

2. *Typical mesoscopic susceptibility*

Combining Eqs. (8) and (11) and multiplying the contributions of the periodic orbits by the corresponding Aharonov-Bohm phases, one finds for the thermodynamic potential of the square billiard

$$\delta\Omega=\frac{2^{5/2}}{\pi^{1/2}}\frac{1}{\Delta}\sum_{p=1}^{\infty}p^{-5/2}\sum_\sigma{}'\left[\frac{\hbar}{T_\sigma}\right]^2\frac{\cos(pk_FL_\sigma-\pi/4)}{(k_FL_\sigma)^{1/2}}\frac{1}{x_\sigma}\int_0^{x_\sigma}dx\,\cos[2\pi pBA_\sigma(x)/\phi_0]\ . \tag{13}$$

The prime restricts the sum over σ to periodic orbits whose contribution depends on the magnetic field. The contribution of each orbit is determined by the average spectral density $1/\Delta$, the factor $(\hbar/T_\sigma)^2$ originating from the energy integral in Eq. (2), and the cylindrical wave amplitude $(k_FL_\sigma)^{-1/2}$. For simplicity, Eq. (13) is shown for zero temperature. However, it should be kept in mind that, strictly speaking, the calculation is valid only for finite temperature or finite inelastic scattering length to satisfy the condition $R_c\gg\min\{L_T,L_\phi\}$. This leads to an exponential suppression of the contributions of long orbits (large L_σ) and of multiple traversals (large p), cf. Eq. (8). The effect of finite temperature is included in the final result (19). Equation (13) yields, for the susceptibility,

$$\delta\chi=\frac{2^{11/2}}{15\pi^{1/2}}\frac{1}{\Delta}\times\sum_{p=1}^{\infty}p^{-1/2}\sum_\sigma{}'\left[\frac{\hbar}{T_\sigma}\right]^2\frac{\cos(pk_FL_\sigma-\pi/4)}{(k_FL_\sigma)^{1/2}}\times\left[\frac{4\pi\mathcal{A}_\sigma}{\phi_0}\right]^2 f(2pB\mathcal{A}_\sigma/\phi_0)\ , \tag{14}$$

where the magnetic-field dependence is described by the function

$$f(\varphi)=30\int_0^1 dx\, x^2(1-x)^2\cos[4\pi\varphi x(1-x)]\ , \quad (15)$$

with $f(0)=1$. The sum over periodic orbits is dominated by the (1,1) orbit and its retracings. The contributions of other orbits are reduced because they are longer and enclose less area. For example, one readily estimates that the contribution of the (3,1) orbit to the susceptibility is approximately 25 times smaller than that of the (1,1) orbit. Thus the typical mesoscopic susceptibility can be approximated by retaining only the (1,1) orbit and its retracings:[29]

$$\langle\delta\chi^2\rangle\simeq\left[\frac{32}{15\pi^{1/2}}\right]^2\frac{1}{\Delta^2}\times\sum_{p=1}^{\infty}\frac{1}{p}\left[\frac{\hbar}{T_{(1,1)}}\right]^4\frac{1}{k_F L_{(1,1)}}\times\left[\frac{2\pi\mathcal{A}_{(1,1)}}{\phi_0}\right]^4 f^2(2pB\mathcal{A}_{(1,1)}/\phi_0)\ . \quad (16)$$

Here the average is taken over an ensemble of systems with different chemical potentials. I assumed that the distribution of chemical potentials is sufficiently wide that the nondiagonal terms in the sum over periodic orbits are suppressed. As it stands, the zero-field susceptibility diverges. However, as emphasized above and in footnote,[29] an exponential cutoff factor of the sum over p due to finite temperature and inelastic scattering is implicit in Eq. (16). Accounting for inelastic scattering in a phenomenological manner, one has roughly

$$\langle\delta\chi^2\rangle\sim\sum_{p=1}^{\infty}\frac{\exp(-2pL_{(1,1)}/L_\Phi)}{p}\approx\begin{cases}1 & \text{if } L_{(1,1)}\approx L_\Phi\\ \ln(L_\Phi/L_{(1,1)}) & \text{if } L_{(1,1)}\ll L_\Phi\ll R_c\ .\end{cases} \quad (17)$$

For the experiment by Lévy *et al.*[1], $L_{(1,1)}\approx L_\Phi$. For this reason only the (1,1) orbit is retained in the following:

$$\langle\delta\chi^2\rangle^{1/2}\simeq\frac{32}{15\pi^{1/2}}\frac{1}{\Delta}\left[\frac{\hbar}{T_{(1,1)}}\right]^2\frac{1}{(k_F L_{(1,1)})^{1/2}}\times\left[\frac{2\pi\mathcal{A}_{(1,1)}}{\phi_0}\right]^2|f(2B\mathcal{A}_{(1,1)}/\phi_0)|\ . \quad (18)$$

The result written in this form is useful because the physical origin of each term remains transparent. It can be expressed more compactly by comparing the amplitude to the bulk Landau diamagnetism $\chi_{\text{Landau}}=-\mu_B^2/3\Delta$ (here $\mu_B=e\hbar/2m$ is the Bohr magneton)

$$\langle\delta\chi^2\rangle^{1/2}\simeq\frac{2^{5/4}}{5\pi^{1/2}}(k_F L)^{3/2}|\chi_{\text{Landau}}|\times\frac{T/T^*}{\sinh(T/T^*)}|f(BL^2/\phi_0)|\ . \quad (19)$$

Here the temperature dependence (8) is included with characteristic temperature

$$k_B T^*=\frac{hv_F}{4\sqrt{2}\pi^2 L}\ . \quad (20)$$

Thus the typical mesoscopic susceptibility is semiclassically larger than the Landau susceptibility and exhibits damped oscillations as a function of the magnetic field as shown in Fig. 3. The magnetic-field dependence of the square billiard is dominated by a single period. The continuous set of areas enclosed by the different periodic trajectories belonging to the (1,1) orbit leads mainly to a damping of the oscillations.

One may ask whether the results change significantly if the high degree of symmetry of the square is broken by deforming it into a rectangle. Clearly, the relevant periodic orbits change continuously from square to rectangle and, hence, qualitatively different behavior is not expected.

3. *Average susceptibility*

The experiment by Lévy *et al.*[1] performed on an ensemble 10^5 squares requires one to calculate the average mesoscopic susceptibility. Whereas the average vanishes when performed in the grand-canonical ensemble, it is nonzero and paramagnetic for small magnetic fields when performed in the canonical ensemble. A straightforward extension[11] of previous work on the average persistent current in the canonical ensemble[10,27] yields

$$\langle\delta\chi\rangle\simeq-\tfrac{1}{2}\Delta\frac{\partial^2}{\partial B^2}\left\langle\left[\int_0^\infty dE\,\delta\rho(E)f_{\langle\mu\rangle}(E)\right]^2\right\rangle\ . \quad (21)$$

A discussion of the limits of validity of this equation in the ballistic regime is presented in Appendix C. In the following this expression is evaluated for the square billiard. For simplicity, the calculation is presented for $T=0$. Finite temperature is reinstated in the final result, Eq. (25).

Using Eq. (11) one finds for the magnetic-field dependent fluctuations of the particle number at fixed chemical potential

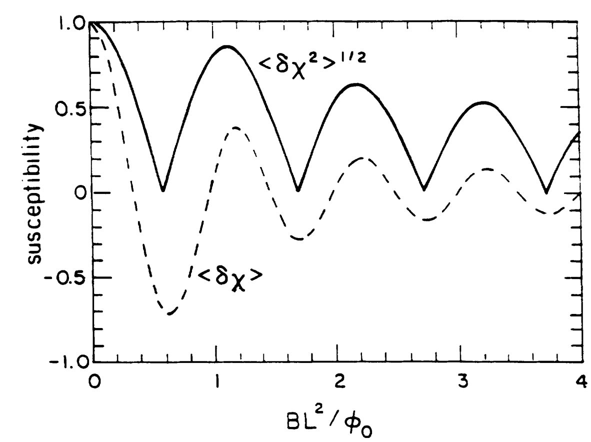

FIG. 3. Magnetic-field dependence of the typical (full line) and the average (dashed line) mesoscopic susceptibility for the square billiard, according to Eqs. (19) and (25), respectively. The susceptibilities are normalized to their values for $B=0$.

$$\delta N=\frac{2^{5/2}}{\pi^{1/2}}\frac{1}{\Delta}\sum_{p=1}^{\infty}p^{-3/2}\sum_{\sigma}{}'\left[\frac{\hbar}{T_\sigma}\right]\frac{\sin(pk_FL_\sigma-\pi/4)}{(k_FL_\sigma)^{1/2}}\int_0^1 dx\,\cos[8\pi pB\mathcal{A}_\sigma x(1-x)/\phi_0]\ . \tag{22}$$

Following the calculation for the typical mesoscopic susceptibility only the contribution of the (1,1) orbit is retained. Combining Eqs. (21) and (22) one has

$$\langle\delta\chi\rangle\simeq\frac{\sqrt{2}}{15\pi}\frac{1}{\Delta}\left[\frac{\hbar v_F}{L}\right]^2\frac{1}{k_FL}\left[\frac{2\pi L^2}{\phi_0}\right]^2 g(BL^2/\phi_0)\ . \tag{23}$$

The function $g(\varphi)$ with $g(0)=1$ describes the magnetic-field dependence

$$g(\varphi)=30\left\{\int_0^1 dx\,\cos[4\pi\varphi x(1-x)]\int_0^1 dx\,x^2(1-x)^2\cos[4\pi\varphi x(1-x)]-\left[\int_0^1 dx\,x(1-x)\sin[4\pi\varphi x(1-x)]\right]^2\right\}\ . \tag{24}$$

In terms of the Landau susceptibility the average mesoscopic susceptibility becomes

$$\langle\delta\chi\rangle\simeq\frac{4\sqrt{2}}{5\pi}(k_FL)|\chi_{\mathrm{Landau}}|\times\frac{(T/T^*)^2}{\sinh^2(T/T^*)}g(BL^2/\phi_0)\ . \tag{25}$$

Here the temperature dependence is included. The magnetic-field dependence is plotted in Fig. 3. Even though the average mesoscopic susceptibility is smaller than its typical value, it is still semiclassically larger than the Landau susceptibility.

4. Comparison with experiment

Lévy *et al.*[1] measured the magnetic susceptibility of an array of 10^5 isolated GaAs squares at subkelvin temperatures. The two experimental samples $S1$ and $S2$ were patterned from high-mobility and high-carrier-density GaAs heterostructures. The squares were 4.5 μm on the side and the elastic mean free path was estimated to be $l_{\mathrm{el}}\approx 5$ μm (10 μm) for $S1$ ($S2$). The electron wavelengths are $\lambda_F\approx 80$ nm (45 nm) for $S1$ ($S2$), and hence $k_FL\approx 350$ (600). I note that the number for k_FL given in the experimental paper is incorrect (assuming that the quoted numbers for L and λ_F are correct). The dispersion in size across the array is 30% (10%) for $S1$ ($S2$). The boundary roughness is estimated from electron micrographs to be less than 50 nm, which is roughly the electron wavelength. The inelastic-scattering length is expected to be between 1.5 and 3 times the elastic mean free path.

The basic observations are as follows. The susceptibility exhibits a paramagnetic zero-field peak whose height is $100\times|\chi_{\mathrm{Landau}}|$ (accurate to within a factor of 2) and whose full width at half maximum is 2.9 Oe, corresponding to roughly one and a half flux quanta threading the square. The peak height decreases with temperature on a scale of 0.5 K.

For the magnetic field $B\approx\phi_0/L^2$, which sets the scale of the experimental field dependence, the cyclotron radius $R_c/L=k_FL/2\pi\approx 60$ (100) for $S1$ ($S2$) is much larger than the linear dimension of the billiard and the phase-coherence length. Hence the results obtained in Sec. II B 3 for the weak-field limit apply. A detailed comparison of the experimental results with Eq. (25) is complicated by the unavoidable fluctuations of the geometry of the experimental "squares" and the residual disorder scattering. In fact, the elastic mean free path quoted in Ref. 1 is shorter than the length of the periodic orbit, on which Eq. (25) is based. Thus the actual periodic orbits of the experimental squares differ from those of the ideal square, and one expects qualitative but not quantitative agreement between experimental and theoretical results. The theoretical prediction Eq. (25) for the amplitude of the average susceptibility in the square billiard is

$$\frac{\langle\delta\chi(B=0)\rangle}{|\chi_{\mathrm{Landau}}|}=\frac{4\sqrt{2}}{5\pi}(k_FL)\approx\begin{cases}140 & \text{for } S1\\ 240 & \text{for } S2\ ,\end{cases} \tag{26}$$

which is slightly larger than the experimental result $100|\chi_{\mathrm{Landau}}|$ (within a factor of 2). This is consistent with the expectation that residual disorder tends to reduce the susceptibility. The magnetic-field dependence is based on the precise area enclosed by the periodic orbits and should be sensitive to deviations from the perfect square geometry. It is reasonable to assume a statistical distribution of the areas enclosed by the dominant periodic orbits of the experimental squares. The corresponding average damps the oscillatory field dependence for the ideal square geometry, cf. Fig. 3, and leads to a single, paramagnetic zero-field peak. The average mesoscopic susceptibility (25) has its first zero at a field corresponding to less than half a flux quantum through the sample. This field is somewhat smaller than the typical field scale of the experimental samples. Thus the dominant periodic orbits of the experimental samples typically enclose less area than the periodic orbit of the ideal square billiard, which is physically reasonable. A similar damping of the magnetic-field dependence due to a distribution of areas should not occur for measurements on single samples, and an oscillatory dependence is predicted by theory.

Whereas the predictions for the amplitude and magnetic-field dependence seem consistent with experiment, the temperature scale is not. From the theoretical result Eq. (25) one finds that the peak height decreases with temperature by a factor of 2 for $T/T^* \simeq 1.5$. For the experimental parameters of Ref. 1 this corresponds to a temperature $T \simeq 0.05$ K which is an order of magnitude smaller than the experimental temperature scale 0.5 K.

C. Integrable and chaotic billiards

The periodic orbits of the square billiard consist of a continuous family of trajectories. This situation is typical of integrable systems. By contrast, almost all periodic orbits of chaotic systems are isolated and unstable. In particular, a system is *completely* chaotic, if *all* periodic orbits are isolated and unstable. An example of such an orbit is shown in Fig. 4 for the Sinai billiard. In this section I derive universal amplitudes of the weak-field susceptibility of integrable and completely chaotic billiard systems. Closely analogous results were recently derived for the persistent current.[22] The results for the susceptibility can be tested more readily experimentally, because it does not require doubly connected geometries threaded by an Aharonov-Bohm flux.

Consider the general periodic-orbit expression for the mesoscopic susceptibility,

$$\delta\chi = \frac{1}{\hbar^\nu} \sum_\gamma A_\gamma \left[\frac{\hbar}{T_\gamma}\right]^2 \left[\frac{2\pi\mathcal{A}_\gamma}{\phi_0}\right]^2 \times \exp\left\{\frac{i}{\hbar} S_\gamma^{(0)}(\mu) + i2\pi B\mathcal{A}_\gamma/\phi_0\right\}. \quad (27)$$

Here $S_\gamma^{(0)}$ denotes the action of the orbit in the absence of a magnetic field. The areas enclosed by the periodic orbits are denoted by $\mathcal{A}_\gamma$. If the different periodic trajectories contained in a nonisolated periodic orbit enclose different areas as for the square billiard, this formula involves an additional integration. I assume that the inelastic-scattering length or the thermal length is of the order of the system size as for the experiment of Ref. 1. Then only the shortest periodic orbits contribute

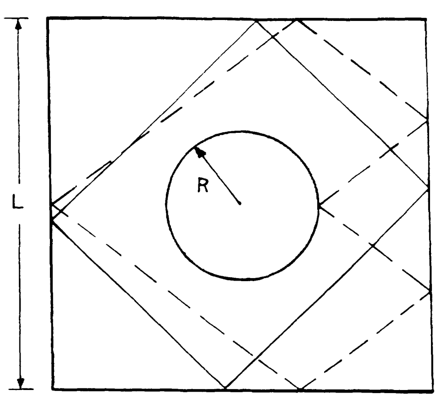

FIG. 4. Sinai-billiard geometry. A marginally stable periodic orbit [$\sigma = (1,1)$] (full line) and an isolated and unstable periodic orbit (broken line) are also shown. The susceptibility exhibits a "semiclassical phase transition" as function of R, since only unstable and isolated periodic orbits contribute to the susceptibility for $R > R_c = L/2\sqrt{2}$. For $R < R_c$ the susceptibility is dominated by the contributions from the marginally stable orbits.

significantly, and it is sufficient to consider the scaling of individual terms in the sum over periodic orbits.

The spectral density of billiard systems satisfies $\rho(E) = (1/\hbar^2) f(E/\hbar^2)$, which implies for the amplitude $A_\gamma(E) \sim E^{(\nu-2)/2}$. Here $\nu = (d+1)/2$ in integrable billiard systems and $\nu = 1$ in completely chaotic ones. Hence for the dependence of the typical mesoscopic susceptibility on the Fermi velocity one finds $\langle \chi^2 \rangle^{1/2} \sim v_F^\nu$. Noting that $\chi_{\text{Landau}} \sim v_F^{d-2}$ (d is the dimensionality of the billiard) this can be written as

$$\langle \delta\chi^2 \rangle^{1/2} \sim |\chi_{\text{Landau}}| \times \begin{cases} (k_F L)^{(5-d)/2} & \text{integrable} \\ (k_F L)^{3-d} & \text{completely chaotic.} \end{cases} \quad (28)$$

Here L denotes a characteristic linear dimension of the billiard. It follows from dimensional analysis that this result fixes the typical mesoscopic susceptibility up to functions of ratios of geometric length scales. If the billiard geometry is defined by a single length scale (e.g., the square or the circular billiard), this expression determines the amplitude up to numerical factors. Analogously, one finds for the average mesoscopic susceptibility in the canonical ensemble that $\langle \delta\chi \rangle \sim v_F^{2\nu-d}$, and hence

$$\langle \delta\chi \rangle \sim |\chi_{\text{Landau}}| \times \begin{cases} (k_F L)^{3-d} & \text{integrable} \\ (k_F L)^{2(2-d)} & \text{completely chaotic.} \end{cases} \quad (29)$$

In the semiclassical limit the mesoscopic susceptibility of integrable billiards is much larger than that of completely chaotic ones. The relative order of magnitude compared to the Landau susceptibility depends on the sample dimensionality. Generally, the mesoscopic susceptibility is more important for smaller sample dimensionalities. Integrable and completely chaotic billiards form only a subset of all possible billiards. One expects that, generically, billiards exhibit mixed dynamics, with both regular and chaotic regions in phase space. The amplitude of the mesoscopic susceptibility of these systems is an interesting open question. Physically one expects that the results for the completely chaotic case also apply to the diffusive regime. Indeed, the scaling (29) of $\langle \delta\chi \rangle$ in completely chaotic billiards is consistent with results obtained for the diffusive regime in Ref. 11.

Nakamura and Thomas[14] inferred from numerical calculations for a family of billiard systems that the deviations from the Landau susceptibility are larger for chaotic than for integrable classical dynamics, in direct contradiction to the result of this paper. These authors use an incorrect scheme to extract the susceptibility from their numerical data. In the integrable limit Nakamura and Thomas[14] calculate the susceptibility separately for each symmetry class of the spectrum, and subsequently add them to obtain the susceptibility of the full system. In this fashion they avoid the numerous level crossings as function of the magnetic field, which, when included, lead to divergences in the susceptibility. Nevertheless, these level crossings must be taken into account. To see this, imagine that a small but finite amount of disorder is introduced into the system, which splits the level crossings. As a result, the level curvature is large but finite. In the limit of zero disorder, the level spacing goes to zero and the level curvature does indeed diverge.

The different contributions to the susceptibility from isolated and nonisolated periodic orbits lead to an interesting "semiclassical phase transition" of the susceptibility in the Sinai-billiard geometry shown in Fig. 4. The classical dynamics of the Sinai billiard is ergodic for any nonzero value of the disc radius R [30]. However, the Sinai billiard is *not* completely chaotic because there are two types of periodic orbits. There are an *infinite* number of isolated and unstable periodic orbits which scatter from the disc, and a *finite* number of marginally stable ones which do not. The marginally stable orbits are remnants of the periodic orbits of the square billiard, and thus they can also be labeled by two integers. Increasingly many of them disappear as the disc radius is increased.[31] The last marginally stable orbit enclosing magnetic flux [the (1,1) orbit] is destabilized at the "critical radius" $R_c = L/2\sqrt{2}$. Therefore I find that due to the existence of marginally stable periodic orbits the persistent current scales as in integrable billiards for $R < R_c$. For $R > R_c$ only isolated and unstable periodic orbits contribute to the susceptibility, and the results for completely chaotic billiards apply. For the typical susceptibility in the semiclassical limit, this implies

$$\langle \delta\chi^2 \rangle^{1/2} \sim |\chi_{\mathrm{Landau}}| \begin{cases} (k_F L)^{3/2}, & R < R_c \\ (k_F L), & R > R_c \ . \end{cases} \tag{30}$$

The analogous result for the persistent current was supported by numerical calculations in Ref. 22.

III. SUMMARY AND DISCUSSION

In this paper the susceptibility of ballistic microstructures has been studied within the independent-electron model. At low temperatures, quantum coherence leads to significant corrections to the thermodynamic limit, if the sample dimensions are of the order of or smaller than the electronic phase-coherence length. Indeed, in this regime the finite-size contributions to the susceptibility studied in this paper can be much larger than the Landau susceptibility.

Semiclassical theory, specifically Gutzwiller's trace formulas, has been used to express the finite-size contributions to the magnetization (mesoscopic magnetization) of ballistic microstructures in terms of the classical periodic orbits. The contribution due to a particular periodic orbit oscillates with chemical potential with the period given by the orbit correlation energy h/T_γ (T_γ is the traversal time of the orbit), and hence the orbit correlation energies determine the scale on which the mesoscopic susceptibility decreases with both temperature and inelastic scattering. The periodic-orbit sum for the mesoscopic susceptibility becomes particularly simple in the weak-field limit, where the cyclotron radius is much larger than the phase-coherence length. In this limit, the periodic orbits can be approximated by those for zero field and the magnetic field enters only through the Aharonov-Bohm phase threading the orbits. In the weak-field limit, the mesoscopic susceptibility is very closely analogous to mesoscopic persistent currents.

Extending previous work on mesoscopic persistent currents I have derived universal dependences of the mesoscopic susceptibility on the Fermi velocity, which fix the amplitude up to functions of ratios of geometric length scales. The mesoscopic susceptibility is much larger for integrable than for completely chaotic billiard systems. This result is in direct contradiction with a claim by Nakamura and Thomas based on numerical calculations for a particular family of billiards. I have shown that these authors eliminate the divergent contributions from level crossings in the integrable case without justification. So far, to my knowledge, no experiment has been performed which measures the mesoscopic susceptibility of a completely chaotic ballistic microstructure.

The mesoscopic susceptibility of the Sinai billiard exhibits a "semiclassical phase transition" between the two universal v_F dependences. For disc radii R smaller than a "critical" radius R_c, marginally stable periodic orbits contribute to the susceptibility and lead to a dependence equal to that of integrable billiards. For $R > R_c$ only isolated and unstable periodic orbits contribute, and the results for completely chaotic billiards apply.

Motivated by a recent experiment by Lévy *et al.* measuring the magnetization of an ensemble of 10^5 GaAs squares, I have studied the mesoscopic susceptibility of the square billiard in detail. The results for amplitude, magnetic-field dependence, and temperature dependence are compared with experiment. The comparison is complicated by the fact that the classical periodic orbit on which the calculation is based is longer than the experimental elastic mean free path. The theoretical results for amplitude and magnetic-field dependence are consistent with experiment, if one invokes the residual disorder scattering to explain that only a single zero-field peak of the susceptibility is seen experimentally. By contrast, a much faster decrease with temperature is predicted than observed experimentally.

The calculation neglects electron-electron interactions, boundary roughness, and residual disorder scattering. By analogy with mesoscopic persistent currents, there may also be a contribution to the ensemble-averaged mesoscopic susceptibility due to interactions. The partial disagreement between theory and experiment emphasizes the need for further theoretical work in this direction. It is interesting to compare the experiment by Lévy *et al.* to the closely related experiment by Mailly, Chapelier, and Benoit[32] measuring persistent currents in a single GaAs ring. While it was also concluded in the latter case that the experimental results for amplitude and field dependence are consistent with the predictions of the independent-electron model, the temperature dependence has not yet been measured.

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APPENDIX A: THE SPIN SUSCEPTIBILITY

In this appendix I estimate the mesoscopic corrections to the spin susceptibility. Motivated by the experiment by Lévy *et al.*[1] it is assumed that the inelastic mean free path L_Φ is of the order of the system size. Denoting the grand potential in the absence of spin interactions as $\Omega_0(\mu)$, one has

$$\Omega(\mu)=\tfrac{1}{2}[\Omega_0(\mu+\mu_B B)+\Omega_0(\mu-\mu_B B)] , \tag{A1}$$

where $\mu_B=e\hbar/2m$ denotes the Bohr magneton. The mesoscopic contributions to the grand potential oscillate as a function of μ with a period of the order of the correlation energy hv_F/L. (Shorter periods are suppressed by inelastic scattering). We can expand to leading order in $\mu_B B$ because

$$\mu_B B=\frac{1}{2}\left[\frac{BL^2}{\phi_0}\right]\left[\frac{hv_F}{L}\right]\frac{1}{k_F L}\ll\frac{hv_F}{L} . \tag{A2}$$

Thus one has

$$\Omega(\mu)\simeq\Omega_0(\mu)-\tfrac{1}{2}\mu_B^2 B^2\rho(\mu) . \tag{A3}$$

The first term leads to the orbital susceptibility. Replacing the density of states in the second term by its average, one recovers the standard expression for the Pauli paramagnetism. The mesoscopic corrections due to the discreteness of the spectrum are

$$\begin{aligned}\delta\chi_{\text{spin}}&=\tfrac{1}{2}\mu_B^2\frac{\partial^2}{\partial B^2}[B^2\delta\rho(\mu)]\\&=\mu_B^2\left[\delta\rho(\mu)+2B\frac{\partial}{\partial B}\delta\rho(\mu)+\tfrac{1}{2}B^2\frac{\partial^2\delta\rho(\mu)}{\partial B^2}\right] .\end{aligned} \tag{A4}$$

In the weak-field limit the magnetic field enters $\delta\rho(\mu)$ only through the Aharonov-Bohm phase factor. Hence one has $\partial\delta\rho/\partial B\sim(L^2/\phi_0)\delta\rho$. For the experiment of Ref. 1 the magnetic field corresponds to roughly one flux quantum threading the sample. As a result, the mesoscopic corrections to the spin susceptibility are of order $\Delta\delta\rho(\mu)$, a quantity which is semiclassically small.

APPENDIX B: THE SPECTRAL DENSITY OF THE SQUARE BILLIARD

The spectrum of the square billiard in the absence of a magnetic field is given by

$$e_{\mathbf{n}}=\frac{\pi^2\hbar^2}{2mL^2}\{n_1^2+n_2^2\} , \tag{B1}$$

where $\mathbf{n}=(n_1,n_2)$ and n_1 and n_2 run from 1 to ∞. The spectral density

$$\rho(E)=\sum_{n_1=1}^{\infty}\sum_{n_2=1}^{\infty}\delta(E-e_{\mathbf{n}}) \tag{B2}$$

can be expressed as a sum over periodic orbits by performing the sum over $\mathbf{n}$ using Poisson summation,

$$\rho(E)\simeq\tfrac{1}{4}\sum_{m_1,m_2=-\infty}^{\infty}\int_{-\infty}^{\infty}dn_1dn_2\exp\{i2\pi(m_1n_1+m_2n_2)\}\times\delta\left[E-\frac{\pi^2\hbar^2}{2mL^2}\{n_1^2+n_2^2\}\right] . \tag{B3}$$

It is not difficult to show (e.g., by direct computation) that the neglected terms make contributions of higher order in Planck's constant $\hbar$. In cylindrical coordinates the integration becomes elementary, and the result can be expressed in terms of the Bessel function $J_0(z)$,

$$\rho(E)=\frac{mL^2}{2\pi\hbar^2}\sum_{m_1=-\infty}^{\infty}\sum_{m_2=-\infty}^{\infty}J_0(kL_{(m_1,m_2)}) , \tag{B4}$$

where L_σ with $\sigma=(m_1,m_2)$ denotes the length of the periodic orbits, cf. Eq. (9), and the wave number k was introduced through $k^2=2mE/\hbar^2$. The well-known average spectral density is recovered by the term $m_1=m_2=0$, $\langle\rho(E)\rangle=1/\Delta\simeq mL^2/2\pi\hbar^2$. All other terms in the sum over (m_1,m_2) oscillate as a function of k. In the semiclassical limit $\hbar\to0$, the wave number becomes large and the Bessel function may be approximated by its asymptotic form. As a result one obtains Eq. (11).

APPENDIX C: THE AVERAGE MESOSCOPIC SUSCEPTIBILITY IN THE CANONICAL ENSEMBLE

In the original works on the average persistent current in the diffusive regime,[10,27] the approximations leading to the analog of Eq. (21) for the average current were made without careful justification. For the diffusive regime, Altland *et al.*[33] showed that these approximations can be circumvented if one averages over both disorder and filling. Here the formula is justified carefully in the ballistic regime, where one averages only over filling.

In the canonical ensemble the magnetization can be obtained from the total energy

$$E(N)=\int_0^{\mu(B)}dE\,E\rho(E) , \tag{C1}$$

where the field-dependent chemical potential is defined by the constant-N constraint

$$N=\int_0^{\mu(B)}dE\,\rho(E) . \tag{C2}$$

Differentiating both $E(N)$ and N with respect to the magnetic field and using that $dN/dB=0$, one obtains the following *exact* formula for the magnetization:[27]

$$M(N)=-\frac{\partial E(N)}{\partial B}=-\frac{\partial}{\partial B}[E(N)-\langle\mu\rangle N]$$
$$=-\tfrac{1}{2}\rho(\mu)\frac{\partial}{\partial B}(\delta\mu)^2-\int_{\langle\mu\rangle}^{\mu}dE(E-\langle\mu\rangle)\frac{\partial}{\partial B}\rho(E)-\int_0^{\langle\mu\rangle}dE(E-\langle\mu\rangle)\frac{\partial}{\partial B}\rho(E)\ . \tag{C3}$$

Here $\langle\mu\rangle$ is defined through the constant-N constraint

$$N=\int_0^{\langle\mu\rangle}dE\langle\rho(E)\rangle\ , \tag{C4}$$

where $\rho(E)=\langle\rho(E)\rangle+\delta\rho(E)$. After performing the average over N (or, equivalently, $\langle\mu\rangle$), the last term in (C3) is negligible because it corresponds to the average mesoscopic susceptibility in the grand-canonical ensemble. Below it is shown that the first term also factorizes to leading order and that the second term is negligible,

$$\langle M(N)\rangle_N\simeq-\tfrac{1}{2}\langle\rho(\mu)\rangle\left\langle\frac{\partial}{\partial B}(\delta\mu)^2\right\rangle\ . \tag{C5}$$

The expression (21) for the average susceptibility follows after relating $\delta\mu$ and $\delta N=\int_0^{\langle\mu\rangle}dE\ \delta\rho(E)$ by means of the constant-N constraint,

$$\int_{\langle\mu\rangle}^{\mu}dE\langle\rho(E)\rangle+\int_0^{\mu}dE\ \delta\rho(E)=0\ . \tag{C6}$$

One may be tempted to conclude that Eq. (21) follows by expanding to first order in $\delta\mu=\mu-\langle\mu\rangle$. However, this expansion is problematic, because by definition $\delta\rho$ has singularities at the (semiclassical) eigenenergies. Hence it is at best a smooth function on scales smaller than the level spacing. Since it turns out that $\delta\mu$ is much larger than the level spacing, we cannot simply expand in Eq. (C6). The way out of this dilemma is suggested by rephrasing the problem in semiclassical language. Semiclassically, $\delta\rho(E)$ is expressed as a sum over periodic orbits. Long orbits give rise to contributions which are only weakly suppressed and which oscillate as function of energy with "frequency" T_N/h. Hence an expansion in $\delta\mu$ breaks down for the contributions of orbits with $T_N>h/\delta\mu$. Their contributions become negligible only if one introduces a cutoff due to inelastic scattering (or finite temperature) such that $\delta\mu\ll h/T_\Phi$. Under this condition, one may expand to leading order in $\delta\mu$, yielding $\delta\mu\simeq-\Delta\delta N$, and hence (21). I stress that the cutoff introduced here is different from that introduced by Altshuler, Gefen, and Imry[10] and Schmid.[27]

The required inelastic scattering cutoff depends on the precise geometry. For the square geometry one estimates

$$\delta\mu\simeq-\Delta\delta N$$
$$\approx\frac{\hbar v_F}{L}\frac{1}{(k_FL)^{1/2}}\ll\frac{h}{T_\Phi}\ . \tag{C7}$$

Thus the phase-coherence length must satisfy $L_\Phi\ll(k_FL)^{1/2}L$. This inequality is satisfied for the parameters of the experiment of Ref. 1.

Now the terms in Eq. (C3), which were neglected in obtaining (C5), can be estimated. Expanding the second term in (C3) in $\delta\mu$, one finds that the leading term before averaging is quadratic in $\delta\mu$,

$$\left\langle\tfrac{1}{2}\delta\mu^2\frac{\partial}{\partial\varphi}\delta\rho(\langle\mu\rangle)\right\rangle\ . \tag{C8}$$

However, after averaging, this contribution is exceedingly small, because the average is over an odd number of oscillating functions. Hence the leading correction after averaging comes from the third-order term,

$$\frac{1}{3}\left\langle\delta\mu^3\frac{\partial}{\partial\langle\mu\rangle}\frac{\partial}{\partial\varphi}\delta\rho(\langle\mu\rangle)\right\rangle\lesssim\langle\delta\mu^2\rangle^{3/2}\frac{T_\phi}{\hbar}\langle\delta\rho^2\rangle^{1/2}\ . \tag{C9}$$

This is much smaller than the leading term (C5) because

$$\langle\delta\mu^2\rangle^{1/2}\frac{T_\Phi}{\hbar}\ll1 \tag{C10}$$

and

$$\langle\delta\rho^2\rangle^{1/2}\ll\langle\rho(\langle\mu\rangle)\rangle\ . \tag{C11}$$

Finally, we need to show that the average of the first term in Eq. (C3) can be factorized. It turns out that corrections to the factorized average are of the same order as (C9),

$$\frac{1}{2}\left\langle\rho(\mu)\frac{\partial}{\partial\varphi}\delta\mu^2\right\rangle=\frac{1}{2}\left\langle\langle\rho(\mu)\rangle\frac{\partial}{\partial\varphi}\delta\mu^2\right\rangle+\frac{1}{2}\left\langle\delta\rho(\mu)\frac{\partial}{\partial\varphi}\delta\mu^2\right\rangle\ . \tag{C12}$$

Hence the corrections to the factorized average are approximately

$$\frac{1}{2}\left\langle\delta\rho(\langle\mu\rangle)\frac{\partial}{\partial\varphi}\delta\mu^2\right\rangle+\frac{1}{2}\left\langle\delta\mu\left[\frac{\partial}{\partial\langle\mu\rangle}\delta\rho(\langle\mu\rangle)\right]\frac{\partial}{\partial\varphi}\delta\mu^2\right\rangle\ . \tag{C13}$$

The first term is vanishingly small because the average is over three oscillating functions. The second is of the same order as (C9). This completes the justification of (C5) under the condition that $T_\Phi\ll h/\langle\delta\mu^2\rangle^{1/2}$.

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