Orbital Magnetism in Ensembles of Ballistic Billiards

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We calculate the magnetic response of ensembles of small two-dimensional structures at finite temperatures. Using semiclassical methods and numerical calculation we demonstrate that only short classical trajectories are relevant. The magnetic susceptibility is enhanced in regular systems, where these trajectories appear in families. For ensembles of squares we obtain a large paramagnetic susceptibility, in good agreement with recent measurements in the ballistic regime.

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A free electron gas at temperature T and magnetic field H such that $k_B T \gg \hbar w$ $(w = eH/mc)$ exhibits a small orbital diamagnetic response [1]. This behavior persists when the electrons are placed in periodic or weak-disorder potentials [2]. When the system is constrained to a finite volume, the confining energy appears as a relevant scale giving rise to finite-size corrections to the Landau susceptibility. These corrections have been the object of several theoretical studies in the last few years for the case of clean [3] and disordered [4] systems, and received renewed interest with recent experiments of Lévy *et al.* [5]: Measurements on an *ensemble* of 10^5 microscopic, phase-coherent, ballistic [6] squares lithographically defined on a high mobility GaAs heterojunction yielded a large paramagnetic susceptibility at zero field, decreasing on the scale of approximately one flux quantum through each square. These experiments have been important in orienting the theoretical studies toward the physically relevant questions associated with the magnetic response of small systems. In particular, the role of finite temperature and the necessity of distinguishing individual from ensemble measurements appear as important ingredients that have been overlooked in some of the theoretical literature.

In this Letter we calculate the orbital magnetic susceptibility of noninteracting electrons at finite temperatures in regular geometries (i.e., squares and circles) for individual systems as well as for ensembles. We use a semiclassical approach treating the magnetic fields involved by classical perturbation theory and confirm the validity of our assumptions and analytical results with numerical quantum calculations. We show that regular microstructures exhibit strongly enhanced susceptibilities with respect to the Landau value due to large modulations in the density of states caused by *families of periodic orbits*. Finite temperature induces a cutoff on the length of the relevant trajectories, and therefore clean systems provide a good description of the ballistic regime. These are the experimental conditions of Ref. [5], and therefore our model yields results in good agreement with the measurements. We compare the results for ensembles of regular geometries with those of chaotic billiards, finding important quantitative differences which should be experimentally observable.

We consider an ensemble of isolated two-dimensional systems at temperature T. For each member of the ensemble (with N electrons and area V) the magnetic susceptibility χ is given by the change of the free energy $F(T,N,H)$ under the effect of a magnetic field,

$$\chi = -\frac{1}{V}\left(\frac{\partial^2 F}{\partial H^2}\right)_{N,T}. \tag{1}$$

The necessity of using the canonical ensemble for isolated mesoscopic systems, and the physical differences with the grand-canonical ensemble (GCE, where the system responds to the magnetic field with a fixed chemical potential μ), are some of the important concepts that recently emerged in the context of persistent currents [7]. On the other hand, calculations in the GCE are more easily performed due to the simple form of the thermodynamic potential

$$\Omega(T,\mu,H) = -\frac{1}{\beta}\int dE\,\rho(E)\ln(1 + \exp[\beta(\mu - E)]), \tag{2}$$

in terms of the single particle density of states $\rho(E) = -(2/\pi)\mathrm{Im}\,g(E)$. The factor of 2 takes into account spin degeneracy, $\beta = 1/k_B T$, and $g(E)$ is the trace of the Green function $G_E(\mathbf{r}',\mathbf{r})$, i.e.,

$$g(E) = \int d\mathbf{r}\, G_E(\mathbf{r},\mathbf{r}). \tag{3}$$

Separating ρ into a mean and an oscillating part, $\rho(E) = \rho^0(E) + \rho^{\mathrm{osc}}(E)$, we define a mean chemical potential μ^0 from $N = \int dE\,\rho(E)f(E-\mu) = \int dE\,\rho^0(E)f(E-\mu^0)$. ($f$ is the Fermi-Dirac distribution function.) Considering that $\rho^{\mathrm{osc}} \ll \rho^0$, it has been shown that [8]

$$F(N) \simeq F^0 + \Delta F^{(1)} + \Delta F^{(2)}, \tag{4}$$

where $F^0 = \mu^0 N + \Omega^0(\mu^0)$ and $\Delta F^{(1)} = \Omega^{\mathrm{osc}}(\mu^0)$. We define Ω^0 and Ω^{osc} by using, respectively, ρ^0 and ρ^{osc} instead of ρ in Eq. (2). The second-order term is [8]

$$\Delta F^{(2)} = \frac{1}{2\rho^0(\mu^0)}\left[\int dE\,\rho^{\mathrm{osc}}(E)f(E-\mu^0)\right]^2. \tag{5}$$

F^0 is field independent to leading order in a semiclassical expansion. Higher order terms in $\hbar$ give rise to the two-dimensional diamagnetic Landau susceptibility $-\chi_{\mathcal{L}} =$

$-e^2/(12\pi mc^2)$ (as can be shown even for constrained geometries [9]). $\Delta F^{(1)}$ gives the susceptibility in a GCE with chemical potential μ^0. In disordered systems it vanishes under impurity average, and we will show that it is also the case within the energy and size averages of our model. We therefore have to consider the next order term $\Delta F^{(2)}$.

To obtain a semiclassical expression for $\Delta F^{(1)}$, $\Delta F^{(2)}$, and their magnetic field derivatives we calculate ρ^{osc} from the semiclassical expansion of the Green function. Except for a logarithmic singularity when $\mathbf{r}' \to \mathbf{r}$, which yields the smooth part ρ^0 of ρ, the semiclassical Green function has the generic form [10]

$$G_E^{\text{sc}}(\mathbf{r}', \mathbf{r}) = \sum_t D_t \exp\left\{ i \left[\frac{S_t}{\hbar} - \left(\eta_t - \frac{1}{2} \right) \frac{\pi}{2} \right] \right\}, \quad (6)$$

where the sum runs over all classical trajectories t joining $\mathbf{r}$ to $\mathbf{r}'$ at energy E. S_t is the action integral along the trajectory. For billiards without magnetic field we simply have $S_t/\hbar = kL_t$, where $k = \sqrt{2mE}/\hbar$ and L_t is the length of the trajectory. The amplitude D_t takes care of the classical probability conservation, and η_t is the Maslov index.

Within our semiclassical approach, the free energy corrections are given as sums over classical trajectories, each term being the convolution in energy of the semiclassical contribution (oscillating as kL_t) with the Fermi factor (smooth on the scale of β^{-1}). It can be seen [9] that the contribution of a given trajectory to $\Delta F^{(1)}$ at finite temperature is reduced with respect to its $T = 0$ counterpart by a multiplicative factor $R(T) = (L_t/L_c)\sinh^{-1}(L_t/L_c)$, with $L_c = \hbar^2 k_F \beta/(\pi m)$. A factor $R^2(T)$ is needed for $\Delta F^{(2)}$. At high temperatures $R(T)$ yields an exponential suppression of long trajectories. Therefore χ is dominated by trajectories with $L_t \leq L_c$, which will be the only ones considered in our analysis. L_c provides a cutoff length in the semiclassical expansion Eq. (6), in a similar way as the phase-coherence length L_Φ associated with inelastic processes. If L_c or L_Φ are much smaller than the shortest classical orbit, χ reduces to the Landau susceptibility independently of the nature of the classical dynamics.

The standard route to obtain ρ^{osc} from G_E^{sc} is to evaluate the integral of Eq. (3)) by stationary-phase approximation. This selects the trajectories which are not only closed in configuration space ($\mathbf{r}' = \mathbf{r}$), but also closed in phase space ($\mathbf{p}' = \mathbf{p}$), i.e., periodic orbits. When these latter are [well] isolated the Gutzwiller trace formula [10] is obtained. For integrable systems, periodic orbits come in continuous families corresponding to the rational invariant tori (Balian–Bloch and Berry–Tabor formulas [11,12]). For regular geometries the $H = 0$ dynamics is integrable. However, χ is the response to a perturbing magnetic field which usually breaks the integrability. Thus using the Berry-Tabor formula is certainly inadequate. On the other hand, for $H \to 0$ the remaining periodic orbits are not sufficiently well isolated to apply the Gutzwiller trace formula. Therefore, a uniform treatment of the perturbing field is needed, where not only orbits that are closed in phase space are taken into account, but also trajectories closed in configuration space which can be traced to periodic orbits when $H \to 0$.

In squares (of side a), due to the simplicity of the geometry, such a uniform treatment is possible since we can perform the corresponding integrals exactly. For $H = 0$, η_t is twice the number of reflections, and $D_t = \alpha/(kL_t)^{1/2}$ with $\alpha = -m/(\sqrt{2\pi}\hbar^2)$. One way to obtain this result is to use the method of images writing $G_E(\mathbf{r}', \mathbf{r}) = G_E^0(\mathbf{r}', \mathbf{r}) + \sum_{\mathbf{r}_i'} \epsilon_i G_E^0(\mathbf{r}_i', \mathbf{r})$, where G_E^0 is the free Green function, $\mathbf{r}_i'$ are the mirror images of $\mathbf{r}'$ by any combination of symmetries across the sides of the square, and $\epsilon_i = \pm 1$ depending on the number of symmetries needed to map $\mathbf{r}'$ on $\mathbf{r}_i'$. The long-range asymptotic behavior of the two-dimensional free Green function $G_E^0(\mathbf{r}_i', \mathbf{r}) \simeq \alpha \exp[i(k|\mathbf{r}_i' - \mathbf{r}| - \pi/4)]/(k|\mathbf{r}_i' - \mathbf{r}|)^{1/2}$ can be used for the images [13].

For sufficiently weak magnetic fields, one may keep in Eq. (6) the zero-order approximation for D_t and use the first-order correction δS to the action. For a closed orbit enclosing an algebraic area $\mathcal{A}$, classical perturbation theory yields $\delta S = (e/c)H\mathcal{A}$ for low fields and high energies, such that the cyclotron radius of the electrons is much larger than the typical size of the structure.

We now specify the contribution ρ_{11} [to ρ^{osc}] of the family of closed trajectories which, for $H \to 0$, tends to the family of shortest periodic orbits with nonzero enclosed area. We note it (1,1) since the trajectories bounce once on each side of the square (upper inset, Fig. 1). Their length is $L_{11} = 2\sqrt{2}a$. This family gives the main contribution to the experiment of Ref. [5] since $L_c \approx 2a$ at $T = 40$ mK. Therefore, to simplify the discussion of the results, we shall in the following also consider that $L_c \leq L_{11}$. We stress, however, that the contributions of other families can be obtained in essentially the same way as the (1,1) contribution. Moreover, strong flux cancellation occurring for other primitive orbits makes their contribution irrelevant in the case of the square, even for very low temperatures [9,14].

In order to calculate the trace integral of Eq. (3) we use as space coordinates x_0, which labels the trajectory (see inset, Fig. 1) and s the distance along the trajectory. Then the area is simply $\mathcal{A}_\epsilon(x_0) = \epsilon 2x_0(a - x_0)$, with the index $\epsilon = \pm 1$ specifying the sense of motion. Inserting $\mathcal{A}$ in Eq. (3) we have $\rho_{11}(H) = \rho_{11}(\mathcal{H} = 0)C(H)$, where $\rho_{11}(\mathcal{H} = 0) = -(8/\pi)a^2\alpha \sin(kL_{11} + \pi/4)/(kL_{11})^{1/2}$ is the unperturbed contribution and

$$C(H) = \frac{1}{a}\int_0^a dx_0 \cos\left(\frac{2e}{\hbar c}Hx_0(a - x_0)\right) = \frac{1}{\sqrt{2\varphi}}[\cos(\pi\varphi)\mathrm{C}(\sqrt{\pi\varphi}) + \sin(\pi\varphi)\mathrm{S}(\sqrt{\pi\varphi})]. \quad (7)$$

C and S are, respectively, the cosine and sine Fresnel integrals, and $\varphi = \Phi/\Phi_0$ is the total flux $\Phi = Ha^2$ inside the square measured in units of $\Phi_0 = hc/e$.

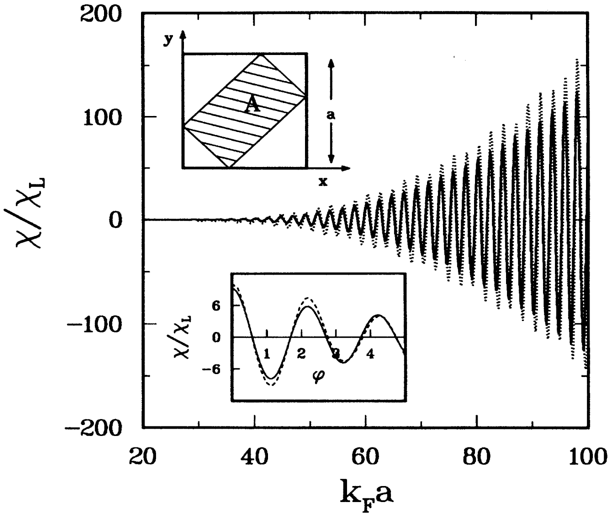

FIG. 1. Magnetic susceptibility of a square as a function of k_Fa at zero field and a temperature equal to 10 level spacings, from numerical calculations (dashed), and from semiclassical calculations (solid). The period $\pi/\sqrt{2}$ indicates the dominance of the shortest periodic orbits enclosing nonzero area with length $L_{11} = 2\sqrt{2}\,a$ (upper inset). Lower inset: amplitude of the oscillations (in $k_F L_{11}$) of χ as a function of the flux through the sample from Eq. (8) (solid) and numerics (dashed).

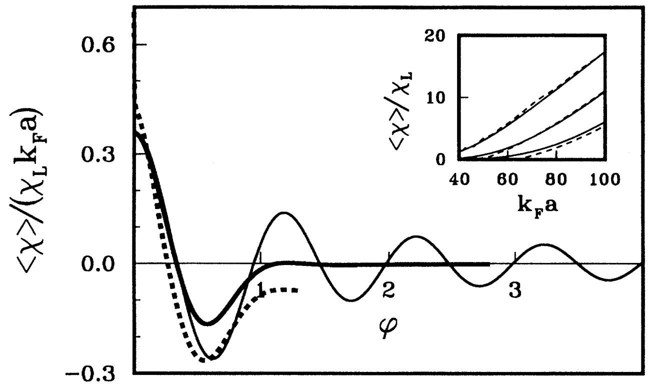

FIG. 2. Thin solid curve: average magnetic susceptibility for an ensemble of squares from Eq. (9). Thick solid curve: average over an ensemble with a large dispersion of sizes (see text). Thick dashed curve: average from numerics. The shift of the numerical with respect to the semiclassical results reflects the Landau susceptibility [due to F^0 in Eq. (4)] not included in the latter. Inset: average susceptibility as a function of k_Fa for various temperatures (4, 6, and 8 level spacings) and a flux $\varphi = 0.15$, from Eq. (9) (solid) and numerics (dashed).

To obtain the contribution of the family (1,1) to $\Delta F^{(1)} = \Omega^{\rm osc}(\mu^0)$ and $\Delta F^{(2)}$ we have to evaluate the energy integrals of Eq. (2) and Eq. (5) using $\rho_{11}(H)$ for the density of states. At $T = 0$, the Fermi distribution is a step function. Since ρ_{11} is a rapidly oscillating function without any stationary-phase point the integrals are dominated by the boundary contribution at the Fermi energy. For finite temperatures the smoothing of the Fermi function results in the factor $R(T)$ previously introduced. For the susceptibility $\chi^{(1)}$ arising from $\Delta F^{(1)}$ one obtains in leading order in k_Fa

$$\frac{\chi^{(1)}}{\chi_L} = \frac{3}{(\sqrt{2}\,\pi)^{5/2}}(k_Fa)^{3/2}\sin\left(k_F L_{11} + \frac{\pi}{4}\right)\frac{d^2C}{d\varphi^2}R(T). \tag{8}$$

Therefore, the susceptibility of a given square can be paramagnetic or diamagnetic (Fig. 1), and its typical magnitude is much larger than the Landau susceptibility χ_L. Clearly, $\chi^{(1)}$ vanishes under average if the dispersion of k_Fa across the ensemble is of the order of 2π. The average χ is then given by the contribution of the (1,1) family to $\Delta F^{(2)}$

$$\frac{\langle\chi\rangle}{\chi_L} = -\frac{3}{(\sqrt{2}\,\pi)^3}k_Fa\frac{d^2C^2}{d\varphi^2}R^2(T). \tag{9}$$

The average susceptibility (solid line, Fig. 2) is paramagnetic at $H = 0$ and for low fields it oscillates with an overall decay of $1/\varphi$. For ensembles with a wide distribution of lengths (in the experiment of Ref. [5] the dispersion in size across the array is estimated between 10% and 30%) the dependence of C on a (through φ) has to be considered. Since the scale of variation of C with a is much slower than that of $\sin^2(k_F L_{11})$, we can effectively separate the two averages and obtain the total mean by averaging the local mean given by Eq. (9). The low-field oscillations of $\langle\chi\rangle$ with respect to φ are suppressed under the second average (performed for a Gaussian distribution with a 30% dispersion, dashed line, Fig. 2), while the zero-field behavior remains unchanged.

We checked the semiclassical results by calculation of the first 1500 eigenenergies of a square in a magnetic field by direct diagonalization. At $T = 0$ the free energy reduces to the total energy and χ is dominated by big paramagnetic singularities at the level crossings of states belonging to different symmetry classes and at small avoided crossings between states with the same symmetry [3]. These peaks are compensated once the next state is considered, and therefore disappear at finite temperature where the occupation of nearly degenerate states becomes almost the same. Temperature regularizes the $T = 0$ singular behavior, and, of course, describes the physical situation. We include it by calculating the partition function $Z = \exp[-\beta F]$ from a recursive algorithm [9,15].

The results for individual squares are in excellent agreement with Eq. (8), the oscillations as a function of $k_F L_{11}$ and φ clearly shown in Fig. 1. The oscillations in φ can be regarded to be analogous to the well known de Haas-van Alphen oscillations of the bulk susceptibility due to quantized electronic cyclotron motion. However, the former explicitly reflect the finite size of the microstructure. At the level of the averages the quantum values also nicely agree with our analytical findings (Fig. 2).

Reference [5] yielded a paramagnetic susceptibility at $H = 0$ with a value of approximately 100 (with an uncertainty of a factor of 4) in units of χ_L. The two electron densities considered in the experiment are 10^{11} and 3×10^{11} cm^{-2}, corresponding to approximately 10^4

occupied levels per square. Therefore our semiclassical approximation is well justified. For a temperature of 40 mK the factor $4\sqrt{2}/(5\pi)k_F a R^2(T)$ from Eq. (9) gives zero-field susceptibility values of 60 and 170, respectively, in good agreement with the measurements. The field scale for the decrease of $\langle\chi(\varphi)\rangle$ is of the order of one flux quantum through each square, in reasonable agreement with our theoretical findings.

Squares constitute a generic example of an integrable system perturbed by a magnetic field. It is interesting to compare our results with two extreme cases: circles (which remain integrable under the perturbation) and completely chaotic systems. Expressing the Hamiltonian of a circle (of radius a) in action-angle variables [16], ρ^{osc} can be written as a sum over families of periodic trajectories [12]. Within our finite-temperature approach we restrict ourselves to the shortest ones, the whispering-gallery trajectories who turn only once around the circle in coming to the initial point after M bounces. Their contribution to ρ^{osc} is

$$\rho_{\text{wg}}(H) = \sum_{M=3}^{\infty} \rho_M(H=0)\cos\left(\frac{eH}{\hbar c}A_M\right). \qquad (10)$$

$\rho_M(H=0) = \sqrt{8}\, m L_M^{3/2}/(\sqrt{\pi}\hbar^2 k_F^{1/2} M^2)\sin(k_F L_M + \pi/4 - 3\pi M/2)$ and the length of the Mth trajectory is $L_M = 2Ma\sin(\pi/M)$, while the enclosed area is $A_M = (Ma^2/2)\sin(2\pi/M)$. The susceptibility $\chi^{(1)}$ oscillates as a function of $k_F a$ with an amplitude proportional to $(k_F a)^{3/2}$ (consistent with Ref. [17]) and vanishes under ensemble average. $\langle\chi(H=0)\rangle/\chi_L \simeq 5.3 k_F a$. The sums over M are rapidly convergent, indicating the dominance of the first few periodic orbits.

Squares and circles give the same dependence on $k_F a$ for $\chi^{(1)}$ and $\langle\chi\rangle$. This is the generic behavior for integrable systems [9] and can be traced to the $(k_F a)^{-1/2}$ dependence of ρ^{osc}. For chaotic systems (of typical length a) with hyperbolic periodic orbits the Gutzwiller trace formula provides the appropriate path to calculate $\rho^{\text{osc}}(E,H)$. For temperatures at which only a few short periodic orbits are important, χ can have any sign, and its magnitude is of the order of $(k_F a)\chi_L$ [18]. Extending this analysis to the case of an ensemble of chaotic systems, we obtain $\langle\chi\rangle \propto \chi_L$. The individual χ are larger, by a factor $(k_F a)^{1/2}$, in regular geometries than in chaotic systems [19]. For $\langle\chi\rangle$ the difference is even of the order of $k_F a$. These differences are due to the large oscillations of ρ in regular systems induced by families of periodic trajectories.

The different magnetic response according to the geometry does not arise as a long-time property (linear vs exponential trajectory divergences) but as a short-time property (family of trajectories vs isolated trajectories). This assures that small variations in the geometry will not be relevant since they affect only long trajectories. For the same reason the effect of weak disorder scattering in the ballistic regime can be treated as a correction to our results for clean systems [9].

The different $k_F a$ dependence predicted for the susceptibility in regular and chaotic cases should result in an order of magnitude effect. Therefore measurements in different geometries will be of high interest and provide a crucial test of the applicability of our noninteracting model to actual microstructures.

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