Nonlinear orbital magnetic response in isolated quantum dots

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The magnetic response of an ensemble of quantum dots all having the same macroscopic parameters but different defect configurations is calculated. The number of electrons in each dot is constant. The response is paramagnetic, can be large at small fields for two-dimensional (2D) dots, and has a characteristic nonlinear behavior including a regime where the magnetization is field independent in 2D, albeit with significant corrections. These results are due to the crossover in the spectral correlations brought about by the time-reversal symmetry breaking due to the magnetic field.

I. INTRODUCTION AND ENUMERATION OF THE VARIOUS FIELD REGIMES

The orbital magnetic susceptibility χ of an electron gas restricted to a finite volume V is still of interest, even for noninteracting electrons. For clean systems, one has the celebrated diamagnetic Landau susceptibility,[1] and various surface or edge corrections may be considered.[2,3] When impurity scattering is included—such that the motion of the electrons becomes diffusive ($L \gg l$, where L is the linear system's size and l is the elastic scattering length)—one still finds that the impurity-averaged susceptibility is (to leading order) equal[4–9] to the Landau one. This holds for the *macroscopic regime* where $L \gg l_\varphi$ or $L \gg l_T$, l_φ denoting the quantum-mechanical coherence length (the "dephasing length") or the inelastic length, and l_T is the thermal length defined by $k_B T = \hbar D / l_T^2$, D being the diffusion constant. In the *mesoscopic regime* ($L < l_\varphi$ and $L < l_T$), the *impurity-averaged* orbital susceptibility is still equal, to leading order, to the Landau diamagnetic susceptibility when the system is held at a fixed field-independent chemical potential (the grand-canonical ensemble).[4–9] Mesoscopic fluctuations are important and have been the subject of numerous recent papers.[4–9]

For noninteracting electrons, it turns out that there is a significant difference between the canonical[10–17] and grand-canonical[18,19] situations (by this we refer to the system being held at a constant number of electrons and chemical potential, respectively, as the magnetic field is varied). An unexpected by-product[10] of the work on persistent currents in mesoscopic rings is the following: Consider the case where χ is averaged over an ensemble of simply connected systems, each of linear size L, characterized by its own microscopic impurity configuration and sharing the same macroscopic parameters (volume, general shape, impurity concentration, etc.). This is the case when a very large system is physically partitioned into many subunits of size L each; one may then measure the magnetic susceptibility of the collection of these subunits. A crossover from a grand-canonical to a canonical situation may be achieved when the ability of the subunits to exchange electrons with a "bath" (e.g., a conducting substrate) is suppressed (e.g., by varying the thickness of an insulating layer separating the system from the bath). It turns out[13,20] that the Coulomb interaction may limit the charge fluctuations and make the situation canonical even in the presence of a coupling to a bath. Denoting these averaged orbital susceptibilities by $\langle \chi_N \rangle$ and $\langle \chi_\mu \rangle$, respectively ($\langle\ \rangle$ stands for impurity-ensemble averaging), one can prove[10,13–15] the following very useful thermodynamic relationship:

$$\langle \chi_N \rangle = \langle \chi_\mu \rangle - \tfrac{1}{2}\Delta \frac{\partial^2}{\partial H^2} \langle \delta N^2 \rangle_\mu , \tag{1.1}$$

where Δ is the average level spacing and $\langle \delta N^2 \rangle_\mu$ is the grand-canonical (fixed μ) average of the subunit-to-subunit fluctuation of the number of particles. This relation is obtained as an expansion of the exact thermodynamic relation $\partial^2 F(T,N,H)/\partial H|_N = \partial^2 \Omega(T,\mu,H)/\partial H^2|_\mu$, neglecting terms of order smaller than Δ/ϵ_F. It will be evaluated below for various regimes employing the Green's-function diagrammatic technique.

Conventional calculations have addressed $\langle \chi_\mu \rangle$. The new term $\langle \chi_N \rangle - \langle \chi_\mu \rangle$ is the main subject of this paper. Evaluating it, one finds—as is originally stated in Ref. 10—a paramagnetic contribution to the orbital susceptibility which can be large, even much larger than the Landau term, in appropriate cases. Some of these results, following Ref. 10, have been obtained parallel with this work in Ref. 8. Here we shall give further results for the nonlinear susceptibility regimes.

It is actually remarkable that the orbital susceptibility is paramagnetic. This is because it measures a profound change in the statistics of the energy levels when a magnetic field or a magnetic flux is applied.[21–26] It is known from the theory of Gaussian random matrices, which describes very well the statistics of levels in a metal on certain scales, that the spectrum becomes more rigid when there is a breakdown of time-reversal symmetry. As a result,

$$\langle \delta N^2(H) \rangle < \langle \delta N^2(0) \rangle \quad \text{hence } \langle \chi_N \rangle - \langle \chi_\mu \rangle > 0 \quad \text{at } H=0 \; . \qquad (1.2)$$

The way this symmetry change shows up in spectral properties (such as $\langle \delta N^2 \rangle_\mu$) will be elaborated upon and used in this paper. In the presence of a flux inside a metallic ring of perimeter L, the effect of the flux has been found at small fluxes to be a function of the unique parameter $E_c \varphi^2$, where E_c is the Thouless energy $\hbar D/L^2$ and φ the dimensionless flux $2\pi\phi/\phi_0$. The change in the spectral rigidity implies that the small energy separations between pairs of levels typically increase with magnetic flux or field. Depending on the number of electrons, one or both members of the pair are occupied. When only one is occupied, it has negative curvature and the small-field susceptibility is paramagnetic. This argument was put forward for the case of the Aharonov-Bohm (AB) flux in Ref. 12. In that case, the flux-dependent part of $\langle \delta N^2 \rangle$ does not exceed an order of 1.

The goal of this paper is to study the average orbital susceptibility of isolated quantum dots. We calculate the paramagnetic contribution $\langle \chi_N \rangle - \langle \chi_\mu \rangle$ for various geometries and eventually we compare it to the Landau diamagnetic susceptibility. We show that the total susceptibility may be paramagnetic in the mesoscopic regime. We limit our study to the diffusive (metallic) regime.

The behavior as a function of the magnetic field is found to be quite intricate. Throughout this discussion, we assume that the relevant linear size of the system, L, satisfies

$$L \ll l_\varphi, l_T \; . \qquad (1.3)$$

Even under this assumption, we identify, in principle, six different regimes for the magnetic response, governed by the following four length scales: (i) The system's size L, (ii) the elastic mean free path l, (iii) the magnetic length $l_H = \sqrt{\hbar c/eH}$, and (iv) the cyclotron radius $l_c = l_H \sqrt{E_F/\hbar\omega_c} \sim k_F l_H^2$; here the cyclotron frequency is $\omega_c = eH/mc$.

The different regimes are depicted in Fig. 1. The weak-field regimes (A, B) are defined by $l_H > L$, i.e., there is less than one flux quantum enclosed in the system. Alternatively this is defined as $H < H_0$, where H_0 is the value of the magnetic field which corresponds to one flux quantum through the system. For the "super-weak-field" case $l_H > (E_c/\gamma)^{1/4} L$ [regime A, $\gamma = \min(\hbar/\tau_\varphi, T)$, where τ_φ is the quantum-dephasing scattering time]. The typical meander of a single-electron energy level as a function of H is smaller than γ, and perturbation theory is applicable as long as $\gamma > \Delta$, Δ being the average level spacing.

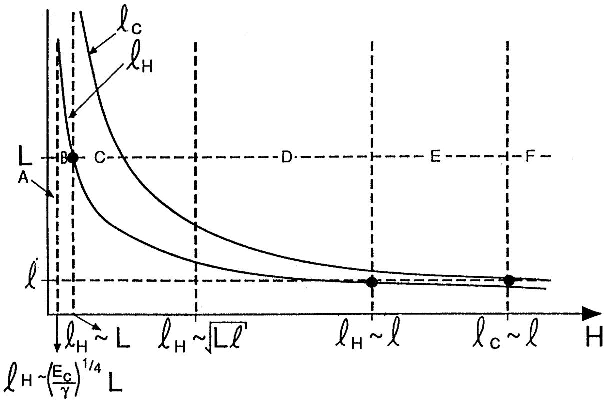

FIG. 1. The various field regimes defined by the characteristic length scales in the problem (for schematic, see text). l_H and l_c are the magnetic length and the cyclotron radius, respectively.

In the mesoscopic regime, $\gamma < E_c$.

The intermediate-field regimes are defined by $L > l_H > l$. This is divided into two subregimes (C, D), as discussed in Sec. III. The strong-field, deep quantum regimes, are defined by $l_H < l$. In regime E, the motion associated with the low-lying Landau levels is not fully diffusive. As we increase the field, more Landau levels fall into this category, until finally one reaches the quantum Hall regime F, where $l_c < l$ or $w_c \tau > 1$ ($\tau = l/v_F$, v_F is the Fermi velocity). (These deep quantum regimes are not treated in this paper.) In the subsequent sections, we study four of these regimes: In Sec. II, we consider the weak-field regimes (A and B) in which nonlinear behavior already appears. We discuss various geometries and point out the importance of the boundaries *even though the motion of the electrons is diffusive.* In Sec. III, we consider the nonlinear $\langle \chi_N \rangle - \langle \chi_\mu \rangle$ in the intermediate-field regimes (C and D). For two-dimensional systems, we find a contribution to the *magnetization* which is field independent. This contribution does not show up in the susceptibility but is of importance for magnetic measurements. The strong-field regimes (E and F) are not treated here.

We recall that for AB geometry, one does not encounter this multiplicity of various regimes. Indeed, for $H < H_0$, the field (or flux) dependence of $\langle \delta N^2 \rangle$ is similar between AB and simply connected geometries. For $H \sim H_0$, $\langle \delta N^2(0) \rangle - \langle \delta N^2(H) \rangle \sim 1$. Due to the flux periodicity in the case of AB flux, this difference remains bound when ϕ is increased. For simply connected systems, this difference increases with H, until finally $\langle \delta N^2 \rangle$ is suppressed by a factor of $\frac{1}{2}$, as is predicted by the general theory of spectral correlations.

II. WEAK-FIELD SUSCEPTIBILITY FOR DIFFUSIVE ELECTRONS

In this section, we evaluate the weak-field susceptibility for diffusive (metallic) systems. As was noted above, we shall assume large enough inelastic broadening ($\gamma > \Delta$, but $\gamma \lesssim E_c$) for our perturbation theory to hold for the very weak-field regime A. From Eq. (1.1) one may express the canonically averaged susceptibility as the sum of the grand-canonical susceptibility and a second term which is paramagnetic (at $H=0$),

$$\langle \chi_N \rangle = \langle \chi_\mu \rangle + \langle \chi_p \rangle , \tag{2.1}$$

with

$$\langle \chi_p \rangle = -\frac{\Delta}{2}\frac{\partial^2}{\partial H^2}\langle \delta N^2 \rangle_\mu .$$

The first term on the right-hand side of Eq. (2.1) is readily shown[4-9] to be the Landau diamagnetic susceptibility χ_L,

$$\begin{aligned} \chi_L &= -\frac{e^2 s k_F}{24\pi^2 mc^2} V \quad \text{in three dimensions} , \\ \chi_L &= -\frac{e^2 s}{24\pi mc^2} S \quad \text{in two dimensions} \end{aligned} \tag{2.2}$$

(we note that in 2D, $\chi_L \sim \Delta / H_0^2$; V and S denote volume and area, respectively). Here k_F is the Fermi wave number, m is the electron's effective mass, and s is the degree of degeneracy of each orbital mode (e.g., due to spin). In the presence of moderate disorder ($1/k_F < l < L$): diffusive regime), the grand-canonically averaged susceptibility $\langle \chi_\mu \rangle$ remains equal to the Landau result (to leading order in $1/k_F l$) for $l_H > l$. As long as the magnetic length is larger than the elastic mean free path, the boundaries of the sample play no important role. A key point of our analysis is that while this statement is valid for thermodynamic, grand-canonically averaged quantities, it does not necessarily apply to fluctuations thereof.

In the present and following sections, we focus on the orbital paramagnetic contribution $\langle \chi_p \rangle$, eventually comparing it with the diamagnetic Landau susceptibility. Our starting point is the calculation of $\langle \delta N^2 \rangle$, using the results of Altshuler and Shklovskii;[21] similar results can be obtained from a semiclassical picture.[24,25] The most divergent contributions to the fluctuations of the particle number are the double-diffuson and double-Cooperon diagrams, shown in Fig. 3 of Ref. 21. These are the most important contributions when dealing with either a large system (summing over a large number of q modes) or a quantum dot (when only the $q=0$ mode is kept, see below).

The diffuson and Cooperon satisfy the equation

$$[-iw - D(\partial_{D,C})^2 + \gamma_{D,C}] P_w^{D,C}(r_1,r_2) = \delta(r_1 - r_2) , \tag{2.3}$$

where γ_D and γ_c represent the inelastic damping of the diffusion and the Cooperon, respectively.

$$\begin{aligned} \partial_D &= \frac{\partial}{\partial r_1} , \\ \partial_C &= \frac{\partial}{\partial r_1} + \frac{2ie\,\mathbf{A}}{\hbar c} , \end{aligned} \tag{2.4}$$

D is the diffusion constant in three dimensions $D = v_F l/3$, and $\mathbf{A}$ is the vector potential associated with the magnetic field. For an isolated system, the boundary conditions of Eq. (2.3) impose zero current across the sample's boundary Σ.

$$n\,\partial_{D,C} P_\omega^{D,C}(r_1,r_2)|_{r_1=\Sigma} = 0 . \tag{2.5}$$

From Eqs. (2.3)–(2.5), it is evident that the diffuson does not depend on $\mathbf{A}$. This is related to the fact that within a semiclassical picture it represents a sum over probability amplitudes of electron trajectories, each multiplied by its complex conjugate. Such terms are insensitive to quantum-mechanical phases. By contrast, the Cooperon contribution correlates trajectories with their time-reversed counterparts. Hence, it consists of squares of probability amplitudes and therefore it is sensitive to $\mathbf{A}$.

The particle fluctuations within an energy range $[-W,0]$ (we shall measure energies from the Fermi energy; W is a high-energy cutoff satisfying $\gamma \ll W \le 1/\tau$) is given by the integral

$$\langle \delta N^2 \rangle_\mu = V^2 \int_{-W}^{0}\int_{-W}^{0} d\epsilon_1 d\epsilon_2 K(\epsilon_1,\epsilon_2) , \tag{2.6}$$

where V is the system's volume. Since we are interested in magnetic-field derivatives of $\langle \delta N^2 \rangle$, we shall consider only the Cooperon contribution to this quantity, $\langle \delta N^2 \rangle^c$. At $H=0$, for a rectangular box geometry,

$$K^{(c)}(\epsilon_1,\epsilon_2) = -\frac{s^2}{2\pi^2 V^2} \,\mathrm{Re} \sum_{\{q\alpha\}} \frac{1}{[\epsilon_1 - \epsilon_2 + i\gamma + iDq^2]^2} , \tag{2.7}$$

where

$$q_\alpha = \pi n_\alpha / L_\alpha , \quad n_\alpha = 0,1,2,\ldots, \quad \alpha = x,y,z . \tag{2.8}$$

In the present section, we treat the magnetic field as a weak perturbation. We note that the term Dq^2 in Eq. (2.7) is obtained as the eigenvalue of the effective Schrödinger equation

$$-D(\partial_C)^2 \Psi_{q_x,q_y,q_z} = E(q_x,q_y,q_z)\Psi_{q_x,q_y,q_z} \tag{2.9}$$

with the boundary condition Eq. (2.5).

The magnetic field gives rise to a quadratic shift of the eigenvalues of the Cooperon: $E_i(H) = E_i + \frac{1}{2}E_i'' H^2$, where $E_i = E_i(0) = D(q_x^2 + q_y^2 + q_z^2)$. The last statement assumes that the set $\{E_i\}$ contains no degeneracy. Starting from Eq. (2.7), it can be shown that

$$\langle \delta N^2 \rangle^c = \frac{s^2}{2\pi^2} \sum_{\{i\}} \ln \left[1 + \left[\frac{W}{\gamma + E_i} \right]^2 \right] , \tag{2.10}$$

where $\{i\} \equiv \{q_i\}$ and $E_i < W$. The precise value of W is irrelevant for the H dependence. Here γ is a cutoff, due either to dephasing of the wave function as discussed in Sec. I or to having a finite temperature. Computing the derivatives of this quantity with respect to the field, we obtain expressions for the paramagnetic moment $\langle M_p(H) \rangle$ and the susceptibility $\langle \chi_p(H) \rangle$.

Special attention should be paid to the mesoscopic regime, when $\gamma \ll E_c = \hbar D / L^2$ (or $L \ll l_\varphi$). Then $E_i \gg \gamma = D / l_\varphi^2$, so that only the $i=0$ mode ($q=0$) is kept in the sum of Eq. (2.10). Consequently,

$$\langle M_p(H)\rangle = \frac{s^2\Delta}{2\pi^2}\frac{E_0''H}{\gamma+\frac{1}{2}E_0''H^2} . \tag{2.11}$$

In the linear, super-weak regime, the magnetization, hence $\langle \chi_p\rangle = (s^2\Delta/2\pi^2)(E_0''/\gamma)$, is *proportional to the diamagnetism of the Cooperon, viewed as a free particle in a box,* of mass 1/2D and charge 2e. The super-weak-field susceptibility is sensitive to the details of the boundary conditions *even in the diffusive regime.*

The magnetization reaches its maximal value at the crossover from regime A to regime B. In the peak (but not super-weak) case $L<l_H<L[E_c/\gamma)^{1/2}(B)$, the magnetization and susceptibility are given, respectively, by

$$\langle M_p(H)\rangle = \frac{\Delta s^2}{\pi^2 H}, \quad \langle \chi_p\rangle = \frac{\partial\langle M_p\rangle}{\partial H} = -\frac{\Delta s^2}{\pi^2 H^2} . \tag{2.12}$$

This is a "universal" result, independent of disorder and geometry. We note that it is also γ independent. Within regime B, the above analysis holds even for $\gamma<\Delta$. The magnetization is parallel to the field, but the curvature is such that the field-dependent susceptibility changes sign and becomes diamagnetic. In 2D, χ is of the order of $\chi_L(H_0/H)^2$.

We now calculate the super-weak-field, linear susceptibility for various geometries. This requires evaluation of the quadratic shifts E_i'' of the Cooperon eigenvalues. There are in principle two different contributions to the field-independent corrections of $E(q_\alpha)$: The first one (the "bulk contribution") arises from the **A** dependence of the operator $(\partial_c)^2$ [cf. Eq. (2.4)]. This will give rise to $O(A^2)$ corrections in $E(q_\alpha)$, calculable within a standard second-order perturbation theory. The second contribution (the "boundary contribution") arises due to the change of the homogeneous Neumann problem (for $\mathbf{A}=0$) to a mixed, Neumann and Dirichlet, boundary-condition problem (for $\mathbf{A}\neq 0$). This second contribution can be calculated using a boundary-condition perturbation theory.[27] The existence of these two contributions, ignored in some previous studies, is evident. Each of them depends on the particular choice of gauge; it is only their sum which is gauge invariant.

The calculation is simplified if only the shift of the ground-state energy of the Cooperon can be considered. We note that this shift does not contain any van Vleck term. This simplification is obtained if we restrict our study to the case where the typical length of the sample in the plane perpendicular to the field is smaller than the phase-coherence length. The length along the field, however, may be either smaller or larger than l_φ. We now calculate $\langle\chi_p\rangle$ for two geometries where the boundary contributions may be ignored. This is the case for a cylinder and a very anisotropic rectangle.

A. Cylinder

The boundary-condition-related contribution can be avoided if one considers a cylindrical geometry with **H** parallel to the cylinder's axis (e.g., $A_r=0$, $A_\theta=\frac{1}{2}Hr$, $A_z=0$). Solutions of the effective Schrödinger Eqs. (2.9) can be parametrized as $\Psi=f(r)e^{im\theta}g(z), m=$integer. The boundary-condition equation reads

$$-i\frac{\partial}{\partial r}f(r)|_{r=R}=0 , \tag{2.13}$$

where R is the cylinder's radius. This equation is independent of H. For $R<l_\varphi$, we need only the shift in the *lowest* eigenvalue which is due only the "bulk contribution" and is equal to $\hbar DR^2/2l_H^4$, as has been found by Altshuler and Aronov.[28] We note, however, that the circular geometry is quite special, as the introduction of a magnetic field does not give rise to matrix elements among the $H=0$ eigenstates (it is analogous to the clean limit in ordinary quantum mechanics, where the above would imply that there are no van Vleck-type contributions to the magnetic susceptibility of the cylindrically symmetric electron system). Throughout this calculation, we recall that $R\lesssim l_H$.

We consider two cases for the cylinder of length L_z.

1. $R<l_\varphi, l_H$; $L_z<l_\varphi$

We include only the $n_z=0$ mode in Eq. (2.10), so that $\langle\chi_p\rangle = (\Delta s^2/2\gamma\pi^2)(\partial^2/\partial H^2)(\hbar DR^2/2l_H^4)$. The paramagnetic correction is written in terms of the Landau susceptibility:

$$\langle\chi_p\rangle = +|\chi_L|\left[\frac{12smDR^2}{\hbar k_F}\frac{\Delta}{\gamma}\frac{1}{V}\right] , \tag{2.14}$$

where $V=\pi R^2 L_z$ is the volume. The level spacing Δ near the Fermi energy is (in 3D)

$$\Delta = \frac{2\hbar^2\pi^2}{smVk_F} .$$

Note that for $\gamma<\Delta$, the factor Δ/γ should be replaced by a constant of order unity which will yield the maximal effect,

$$\frac{\langle\chi_p\rangle}{|\chi_L|}\sim\frac{4s}{\pi}\frac{l}{L_z} . \tag{2.15}$$

We note that this ratio can reach values exceeding unity. In the strict two-dimensional (2D) limit, $L_z\sim k_F^{-1}\ll l$, it reaches a maximum value of the order of $k_F l\gg 1$.

For $\gamma>\Delta$, the result Eq. (2.14) may also be cast in the form

$$\langle\chi_p\rangle = |\chi_L|\frac{24}{(k_F L_z)^2}\left[\frac{l_\varphi}{R}\right]^2 . \tag{2.16}$$

We see that as we decrease γ, the total susceptibility becomes larger. For a two dimensional sample ($k_F L_z\simeq 1$), it reverses sign for $\gamma\sim D/R^2$ and reaches a positive value of order:

$$\langle\chi_p\rangle/|\chi_L|\sim\frac{\hbar D/R^2}{\Delta} \tag{2.17}$$

for $l_\varphi>\sqrt{\hbar D/\Delta}$ [in that limit, l_φ in Eq. (2.17) is replaced by $\sqrt{\hbar D/\Delta}$].

2. $R < l_\varphi, L_H$; $L_z > l_\varphi$

Now we have to sum over the n_z modes, all having the same shift in H. The summation is transformed into an integral $\Sigma_{qz} \to L_z/\pi \int dq_z$. As a result, the paramagnetic term is *enhanced* by a ratio L_z/l_φ compared with Eq. (2.16), so that

$$\langle \chi_p \rangle \sim |\chi_L| \frac{1}{(k_F R)^2} \frac{l_\varphi}{L_z} . \quad (2.18)$$

This corresponds to the addition of L_z/l_φ contributions. Each piece of length l_φ along z contributes independently.

B. Rectangle

For a sample of the square cross section, one needs to consider the finite contribution of the boundary term to second-order corrections to the eigenvalues. We have also to include a mixed contribution, resulting from calculating the first-order correction to the operator $(\partial_c)^2$ with the first-order-corrected eigenfunctions due to the change in the boundary conditions. In the following discussion, we choose an anisotropic rectangular geometry such that, subject to an appropriate gauge, the boundary contribution may be neglected. We should note, however, that as the limit of a square cross section is approached, this boundary contribution introduces an *extra* field-dependent contribution into the Cooperon, on the order of the contribution calculated below.

We consider the anisotropic geometry shown in Fig. 2, where $L_x \ll L_y$. The magnetic field is perpendicular to the xy plane. (Within the linear response, one may deal with each field component separately.) The gauge selected is

$$\mathbf{A} = (0, -Hx, 0) ,$$

leading to a correction in the eigenvalue equation (2.3). We now use Eq. (2.10) where, choosing $L_x \ll l_H, l_\varphi$, the sum $\{i\}$ is performed over the q_y, q_z modes. In this case, it is found that the shift in the eigenvalues is[28] $Dq^2 \to Dq^2 + DL_x^2/3l_H^4$.

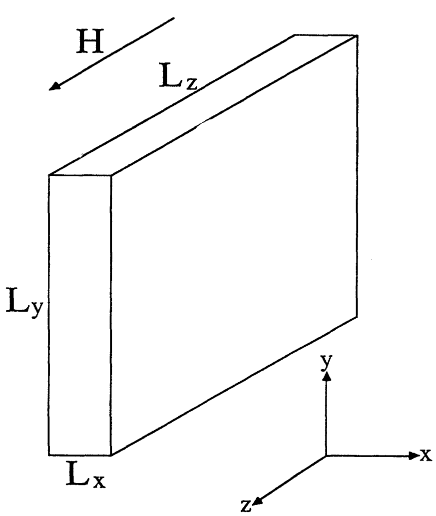

FIG. 2. The perpendicular field in a rectangular geometry ($L_x \ll L_y$).

Note that with the gauge $\mathbf{A} = (Hy, 0, 0)$, the L_x^2 factor should be replaced by $L_y^2 (\gg L_x^2)$ in the H-dependent term of the energy shift. But with that gauge, the boundary contribution is no longer negligible, and will compensate for most of the $DL_y^2/3l_H^4$ term.

In the simplest limit of our rectangular geometry, $L_x, L_y, L_z \ll l_H, l_\varphi$, the shift of the lowest eigenvalue is $DL_x^2/3l_H^4$ instead of $DR^2/2l_H^4$ in the cylinder geometry. The results we obtain are similar. We find

$$\langle \chi_p \rangle = |\chi_L| \frac{16\pi^2}{(k_F L_z)^2} \left[\frac{l_\varphi}{L_y} \right]^2 . \quad (2.19)$$

III. INTERMEDIATE FIELD: DIFFUSIVE REGIME

We now study the limit $l < l_H < L_x, L_y$ (i.e., regimes C and D, cf. Fig. 1). For the sake of concreteness, we consider a rectangular box. For regime D, it turns out that, in contrast to the super-weak-field limit, the details of the sample's geometry do not play a crucial role. For this regime, when $L_z \ll l_\varphi$, the Cooperon correlation function [cf. Eq. (2.7)] assumes the form

$$K^C(\epsilon_1 - \epsilon_2, H) \cong -\frac{eHs^2}{2\pi^3} \frac{L_x L_y}{V^2} \times \operatorname{Re} \sum_{n=0}^{n_m} \frac{1}{[\epsilon_2 - \epsilon_1 + 4ieDH(n+\frac{1}{2})]^2} , \quad (3.1)$$

where the summation is over the Landau levels of the effective Schrödinger equation with a cutoff

$$n_m = \frac{l_H^2}{2l^2} . \quad (3.2)$$

(This is based on the fact that, for eigenvalues larger than $\sim 1/\tau$, the form of the Cooperon should be modified and the related contribution may be neglected.) In this intermediate regime, the vector potential appears as a phase factor in the single-particle eigenfunctions. It may be ignored in the expressions for the single-particle Green's functions.

The classical radius corresponding to the highest Cooperon Landau level, n_m, is

$$R_{n_m} \sim \frac{l_H^2}{l} . \quad (3.3)$$

In regime D ($Ll > l_H^2 > l^2$), this radius is indeed smaller than the linear system's size. In regime C ($L^2 > l_H^2 > Ll$), this is no longer the case, and one needs in principle to sum separately over Landau levels with a cutoff $n_m \sim L^2/l_H^2$, and higher-lying "box-states," which are the original $H=0$ states weakly perturbed by the magnetic field.

Before discussing the results of more careful evaluations based on the above, we try to reach a qualitative understanding of the behavior of $\langle \delta N^2(H,W)\rangle$ as a function of H. We note that for $H=0$, performing the integrations of $K(\epsilon_1-\epsilon_2)$ to obtain $\langle \delta N^2(0,W)\rangle$, one finds that they are dominated by the high-energy parts[21] (this is due to the power-law nature of K and to the cancellations of the lower-energy contributions). For $E_c \ll W \leq h/\tau$, $\langle \delta N^2(0,W)\rangle \propto W^{d/2}$. For H in the intermediate regime, the Cooperon contribution to $\langle \delta N^2(H,W)\rangle$, arising from the energy range $|\epsilon| \ll \hbar\omega_H$, is much smaller than the H-insensitive diffuson contribution; it is approximately equal to the latter for $\epsilon \gg \hbar\omega_H = 4eDH$. The quantity $\hbar\omega_H$ is the crossover energy[21,25] in this regime between orthogonal and unitary behavior. Thus one expects[25] that, in the intermediate regime in d dimensions,

$$\langle \delta N^2(0,W)\rangle - \langle \delta N^2(H,W)\rangle \sim \tfrac{1}{2}\delta N^2(0,\hbar\omega_H) \sim \left[\frac{H}{H_0}\right]^{d/2} \qquad (3.4)$$

(similar considerations can also be made in the weak-field regime). This result indeed follows to leading order from more detailed evaluations. Below we give the calculation for the interesting case of $d=2$. We point out that from Eq. (3.4) it follows that the magnetization is proportional to $H^{d/2-1}$ and the susceptibility to $H^{d/2-2}$. In 2D, one obtains the unusual result that $\langle M_p\rangle$ is a universal constant *independent of* H and disorder, and the field-dependent paramagnetic susceptibility $\langle \chi_p\rangle \equiv \partial\langle M_p\rangle/\partial H$ is zero to leading [see (3.7)] order. However, an examination of the Cooperon approximation [(3.1)] reveals significant deviations from the result [A. Altland and P. Gefen (unpublished)].

The leading approximation of the behavior in regime D is obtained by dropping the cutoff, Eq. (3.2), and neglecting all corrections in the denominator of Eq. (3.1). The sum is easily evaluated in terms of derivatives of Ψ and Γ functions; hence, the integrals are straightforward, and we find, for the Cooperon contribution,

$$\langle \delta N^2\rangle^c = \frac{s^2}{8\pi^2}\frac{W}{E_c} - \frac{eHs^2}{2\pi^3}S(\ln 2 - e^{-2\pi W/\omega_H}) . \qquad (3.5)$$

Apart from the exponentially small corrections, we have an H-independent part which agrees with Ref. 21, and a linear H-dependent part, as expected. It is more difficult to perform an accurate calculation in regime C; a numerical evaluation of Eq. (3.1), including the high-lying box states, is needed. However, since the sum is dominated by $n \sim \epsilon_1 - \epsilon_2/\omega_H$, and the relevant values of $\epsilon_1-\epsilon_2$ are up to a few times $\hbar\omega_H$, we expect the results to be qualitatively the same as in range D. We also note (see below) that the extrapolated results on the D-C and B-C boundaries are of the same order of magnitude, which is consistent with the whole region C behaving similarly to D. Equation (3.5) leads to a paramagnetic H-independent moment (here we restore the $\hbar$ and c factors)

$$\langle M_p(H)\rangle = \frac{e\Delta s^2/\ln 2}{4\pi^3\hbar c}S = \frac{s^2\Delta}{2\pi^2 H_0}\ln 2 \sim |\chi_L| H_0 . \qquad (3.6)$$

This is "universal," in the sense of being disorder independent and sensitive only to the effective mass or density of states. We note again that extrapolating this to the edge of the intermediate-field regime, $H\sim H_0$, agrees with the weak-field result Eq. (2.12), extrapolated to $H \sim H_0$ as well.

The constant M yields a vanishing susceptibility $\partial M/\partial H$. The corrections to the latter can be obtained by inserting the corrections to the "Landau levels" due to the boundaries and the upper cutoff, Eq. (3.2), in Eq. (3.1). In regime D, this yields the two respective corrections

$$\langle \chi_p\rangle \sim |\chi_L| \frac{l_H^2}{k_F L_z L_x L_y}\left\{\alpha\left[\frac{l_H}{L}\right]^2 + \beta\left[\frac{l}{l_H}\right]^2\right\} , \qquad (3.7)$$

α and β being numerical coefficients. These two terms can be regarded as coming from the smaller and larger field regimes, respectively. It appears that to leading order, χ and M are "universal" in the entire weak (B) and intermediate ($C+D$) regimes.

IV. DISCUSSION

We have demonstrated that the zero-field orbital susceptibility in the metallic (diffusive) regime contains a paramagnetic contribution $\langle \chi_p\rangle$ which, in the mesoscopic regime, saturates to

$$\langle \chi_p\rangle \sim |\chi_L| \frac{l}{L_z} \quad \text{cylinder} \qquad (4.1)$$

$$\langle \chi_p\rangle \sim |\chi_L| \frac{l}{L_z}\frac{L_x}{L_y} \quad \text{anisotropic rectangular box } (L_x \ll L_y) . \qquad (4.2)$$

We expect that for any cross section which is not too anisotropic, χ_p is given (up to a numerical factor) by Eq. (4.1).

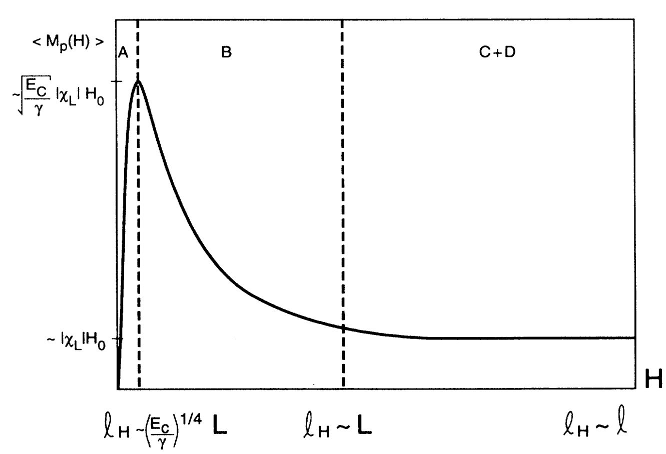

FIG. 3. The nonlinear paramagnetic magnetization $\langle M_p(H)\rangle$ in the weak- and intermediate-field regimes (schematic).

Strong nonlinear dependences of the magnetization and susceptibility are obtained in the weak but not super-weak regime B [see Eq. (2.12)], and in the intermediate regimes $C+D$ [see Eq. (3.6)]. They are schematically depicted in Fig. 3 for 2D. They are universal, i.e., disorder independent (as well as dimensionality independent in regime B), and particularly interesting in the 2D intermediate-field regime. The independence on disorder suggests the interesting question of whether some of those results should hold in the ballistic chaotic regime.

We also note that our results suggest that the dependence of the susceptibility on system's size may not be monotonic: in the macroscopic ($l_\varphi \ll L$) regime $\chi \simeq -|\chi_L|$, the system is diamagnetic, its susceptibility being extensive in the volume. For atoms, one expects Larmor diamagnetism (which is superextensive) to dominate. The crossover between these two regimes goes through a mesoscopic paramagnetic regime, where the susceptibility is extensive and subextensive in $d=2$ and 3, respectively. The experimental data of Refs. 29 and 30 may support this picture. However, more experimental evidence is needed. The additional effect of electron-electron interactions[8,13,31] on the susceptibility is of interest as well.

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