Sign Reversals of ac Magnetoconductance in Isolated Quantum Dots

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We have measured the electromagnetic response of micron-size isolated GaAs/GaAlAs square dots down to 16 mK by coupling them to a microresonator. Both dissipative and reactive responses exhibit a large magnetic field dependent quantum correction, with a characteristic flux scale corresponding to a flux quantum in a dot. The dissipative magnetoconductance changes sign as a function of frequency for a low density of electrons. The signal observed at a frequency below the mean level spacing corresponds to a negative magnetoconductance, in contrast to weak localization in connected systems, and becomes positive at higher frequencies. We interpret this phenomenon in relation to the energy spectrum statistics in the dots. [S0031-9007(98)06250-4]

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There are several ways of measuring the electrical conductance of a metallic sample. The most common one is to drive a current through it and measure the voltage drop between two contacts. However, it is also possible to measure the conductance of the same object without making any connections, via capacitance and inductance measurements at finite frequency. When a metallic sample of typical size a is inserted into a coil, it acquires a magnetic moment \( m = Ia^2 \), where \( I \) is the induced eddy current. This changes the impedance of the coil by a quantity:

\[ \delta Z(\omega) = L \omega \chi_m(\omega)/V, \]

where \( V \) is the volume of the coil of inductance \( L \) and \( \chi_m \) is the magnetic susceptibility of the sample related to its conductance \( G_m \) (the ratio between \( I \) and the induced electromotive force) by [1]

\[ \chi_m(\omega) = i \omega \mu_0 G_m(\omega)a^4. \] (1)

Similarly, the change in admittance of a capacitance \( C \) due to the insertion of a metallic sample reads \( \delta Z^{-1}(\omega) = C \omega \chi_e(\omega)/V \), where \( V \) is the volume of the capacitor and \( \chi_e(\omega) \), the electrical polarizability of the sample, is related to its effective conductance \( G_e \) (defined as the ratio between polarization current and unscreened voltage drop), by [2]

\[ \chi_e(\omega) = \frac{G_e(\omega)a^2}{i \omega \epsilon_0}. \] (2)

In the classical limit at low frequency (such that \( a \) is much smaller than the skin depth) \( G_m \) is a real quantity, identical to the Drude conductance \( G_D = \sigma a \) within a numerical constant which depends on the geometry. On the other hand, due to screening of the electric field inside the metal [3], and for \( \omega < \sigma/\epsilon_0 \), \( G_e \) is dominated by its (reactive) imaginary part \( \text{Im} G_e = \omega a \epsilon_0 \), while its (dissipative) real part \( \text{Re} G_e = (1/\epsilon_0 \omega a^2)/G_D \).

In a mesoscopic object, which has dimensions smaller than the electronic phase coherence length, it has been shown theoretically [4–8] that the conductance depends strongly on the way it is measured. More precisely, it is very sensitive to the coupling between the mesoscopic system and the classical macroscopic apparatus. When a sample is connected with wires, (i.e., when the coupling is strong), relative quantum corrections to the conductance are of the order of \( 1/g \) where \( g \) is the dimensionless Drude conductance [9]. This result concerning the dissipative components of the conductance is unchanged for unconnected samples, provided that the energy level spectrum is continuous [7], i.e., the typical level width \( \gamma \) is much larger than the mean level spacing \( \Delta \). Note however that the quantum corrections of \( \text{Re} G_e \) \( \approx 1/G_D \) and \( \text{Re} G_m \approx G_D \) are opposite in sign. Furthermore, \( \text{Im} G_m \) is expected to be finite and directly related to persistent currents and more generally orbital magnetism in the sample [4–6,10]. At the same time \( \text{Im} G_e \) acquires a magnetic field dependence related to quantum corrections to the electrical polarizability [11]. On the other hand, if the discrete spectrum limit \( \gamma \ll \Delta \) is achieved, quantum coherence gives rise to giant magnetoconductance [6–8], the sign and amplitude of which are determined by the ordering of the relevant energy scales: \( h \omega, \gamma, \Delta, 5k_BT \). Preliminary experiments [12] on an array of Aharonov-Bohm rings coupled to a resonator have shown that the investigation of ac conductance on isolated samples in the discrete spectrum limit is indeed possible.

In this Letter, we report the measurement of the electromagnetic response of an array of mesoscopic square dots made from a GaAs/GaAlAs 2D electron gas at frequencies both below and above the energy level spacing. Sign reversals of the dissipative magnetoconductance were observed and can be interpreted in relation to the sensitivity of level spacings distribution in the dot to magnetic field, according to random matrix theory.

The mesoscopic samples are coupled to an electromagnetic microresonator, made of two Nb wires (\( d = 4 \mu m \) spaced, 1 \( \mu m \) width and 20 cm long) deposited in a meandering geometry on a sapphire substrate [12]. The superconducting material is used to reduce losses and therefore achieve a high quality factor (80 000 with no sample). The resonance conditions are as follows: \( L = \frac{na^2}{2} \), where
$n = 1, 2, 3, \ldots, L$ is the length of the superconducting wires, and $\lambda$ the electromagnetic wavelength. The fundamental resonance frequency is of the order of 330 MHz. The resonator is enclosed in a metallic box, protecting it from electromagnetic noise, and cooled down to 16 mK with a dilution fridge. This experiment allows us to detect extremely small variations of frequency and quality factor: \[ \frac{\delta f}{f} \sim \delta(Q^{-1}) \sim 10^{-9}. \] When mesoscopic samples are deposited on the top of the resonator (with a 1 \( \mu \)m thick mylar sheet spacer), its characteristics are modified. Dissipation in the samples affects the quality factor, whereas the reactive (nondissipative) response affects the resonance frequency, according to

\[
\frac{\delta f}{f} = -kN_s\chi'(\omega) \quad \text{and} \quad \delta(Q^{-1}) = kN_s\chi''(\omega),
\]

where \( \chi(\omega) = \chi_e(\omega) + \chi_m(\omega) \) is the average complex susceptibility of a dot, \( N_s \) is the number of dots, and \( k \) is a geometrical coupling coefficient which is of the order of \( 1/Ld^2 \), where \( d \) is the distance between the two niobium wires. As a consequence, \textit{a priori} our experiment cannot be reduced to a pure capacitance or a pure inductance measurement. Rather, it measures the total response (reactive and dissipative) of a sample to an electromagnetic excitation. Nevertheless the response of the sample itself can be dominated by one of the two components, electric or magnetic. As a matter of fact, we will show later that here, we are dealing with (orbital) magnetic response, i.e., our data correspond to \( \chi_m \) or \( G_m \).

The sample studied in this experiment is an array of 10^5 disconnected square dots etched in a GaAs/GaAlAs semiconductor heterojunction using electron beam lithography techniques. The electronic density of the 2D electron gas (2DEG) before etching was \( n_e = 3 \times 10^{11} \) cm\(^{-2}\). \( n_e \) is strongly depressed in etched samples but can be recovered under illumination. For this purpose an electroluminescent diode was placed next to the resonator, allowing us to gradually change the number of electrons in the squares by transient illumination of the samples (between 30 s and 2 min). The measurements were always done at least one hour after the illumination was stopped, in order to ensure good stability of the samples. The mean level spacing in one square \( \Delta = 2\hbar^2\pi/m^*a \) (\( m^* \) is the effective electron mass) is independent of the electronic density. It is of the order of 38 mK which corresponds to a frequency of 760 MHz. The elastic mean free path of the 2DEG was \( l_e = 10 \mu \)m. From transport measurements and study of weak localization on long wires made together with the present squares, similar to experiments depicted in [13], one could estimate the size of the squares \( a = 1.5 \mu \)m and assure that \( l_e \) is not very different from its value in the unetched heterojunction. As a result the transport is ballistic in the dots.

We have measured the magnetic field dependence of the resonance frequency and quality factor for several harmonics (\( f_1 = 330 \) MHz, \( f_2 = 670 \) MHz, \( f_3 = 1065 \) MHz) and after successive illuminations. The dc magnetic field was modulated at low frequency (~30 Hz) with a small coil close to the sample. This allows measurement of derivatives of the resonance frequency (see Fig. 1) and quality factor with respect to the magnetic field. The signal consists of steplike features on the field dependence of \( \frac{\delta f}{\delta H} \). The contribution of the empty resonator (linear in magnetic field) has to be subtracted from the signal. After integration, the magnetococonductance of the samples \( \delta G(H) \) is \( \chi'(H) - \chi(0) \) can be deduced from these measurements (see Fig. 2). The real and imaginary components of \( \chi \) are of the same order of magnitude and exhibit the striking triangular shape observed in experiments on connected ballistic dots of similar geometries. This feature is believed to be related to the classical integrability of these systems [14].

\[
\delta \chi'(H) \quad \text{and} \quad \delta \chi''(H)
\]

are found to have very different behaviors as a function of frequency and electronic density. \( \delta \chi'(H) \) is always positive; its amplitude drastically increases with the first successive illuminations (which correspond to an increasing number of electrons) and then tends to saturate. For a fixed number of electrons it is nearly independent of frequency [15]. On the other hand \( \delta \chi''(H) \) is observed to be negative for the first harmonics and small electronic density and becomes positive with increasing frequency and electronic density. These singular sign reversals of the dissipative magnetococonductance with frequency and number of electrons are unique properties, characteristic of the isolated geometry of our experiment. They constitute the most striking result of our work.

The temperature dependence of the signals were also measured. The temperature decay depicted in Fig. 3 is

![FIG. 1. Field derivative of resonance frequency for the first harmonics of the resonator. The different curves correspond to successive illuminations of the sample. Inset, bottom: Schematic picture of the resonator and the square dots. Inset, top: Data obtained after subtraction of the empty resonator contribution.](image-url)
found to be independent of the frequency and nearly identical for $\delta \chi'$ and $\delta \chi''$. It cannot be fitted with an exponential as was the case for the ring experiment. The characteristic temperature scale of this decay increases slightly with the number of electrons in the dots.

In the following we compare our results with theoretical predictions on orbital magnetism and polarizability. From Eqs. (1) and (2) one can calculate the ratio $r = \chi''_m / \chi''_e$ between classical magnetic and electric losses. This is given by $r = (Z_0 G_D)^2$, where $Z_0 = \frac{\epsilon \mu_0}{\sqrt{\mu_0 \epsilon}} \approx 377 \, \Omega$ is the vacuum impedance. In our case $r$ varies between 8 and 400 depending on the electronic density in the dots [16]. This result can be shown to remain valid for the field dependent response of phase coherent systems [17]; i.e., $\delta \chi'_m / \delta \chi'_e = \delta \chi''_m / \delta \chi''_e = r$. Therefore, we conclude that the signals observed in the squares are dominated by the magnetic response $\chi_m$, in contrast to the case of the rings previously studied. This fact is confirmed by the observation of an increase of the signals with electronic density (electrical response $\chi_e$), as expected for a discrete level system.

We now discuss in turn the reactive and dissipative responses. The amplitude of $\delta \chi'(H)$ corresponds to an average orbital susceptibility of the order of the theoretical prediction: $g a^2 \mu_0 e^2 / m$ [18]. However, whereas the theory predicts paramagnetism in zero field, in agreement with dc experiments [19], on the other hand our results unambiguously point to diamagnetism. Orbital diamagnetism of similar magnitude has also been recently observed for the persistent currents in mesoscopic rings made of gold [20] and does not have yet a theoretical explanation.

The field dependence of the dissipation of Aharonov-Bohm rings in a time dependent flux has been extensively studied [4–7]. It is straightforward to extend these results to singly connected geometries and estimate the relevant quantity $\text{Re} \, \delta G_m(H) \propto \delta \chi''_m(H) / \omega$. There are two different contributions to the dissipative conductance $\delta G_{\text{off}}$ and $\delta G_{\text{di}}$ in the limit where $\gamma \ll \Delta$ and $T > \Delta$. $\delta G_{\text{off}}$ is related to interlevel transitions and is expressed in terms of the level spacing distribution function $R(s)$, which obeys the universal rules of random matrix theory [21,22]. For $H = 0$, it corresponds to the Gaussian orthogonal ensemble (GOE), and, for sufficiently large field, where time reversal symmetry is broken, to the Gaussian unitary ensemble (GUE).

$$\delta G_{\text{off}}(H) = G_D \int \frac{\gamma[R_{\text{GUE}}(s) - R_{\text{GOE}}(s)]}{(\hbar \omega - s)^2 + \gamma^2} \, ds. \quad (4)$$

On the other hand, $\delta G_{\text{di}}$ is related to the relaxation of persistent currents and always yields a positive magnetoconductance:

$$\delta G_{\text{di}}(H) = G_{\text{di}}(\text{GUE}) - G_{\text{di}}(\text{GOE}) = G_D \frac{\gamma \Delta}{\omega^2 + \gamma^2}. \quad (5)$$
In the limit where \( \gamma \ll \omega \) the magnetoconductance is dominated by \( \delta G_{m}(H) \sim G_{D}[R_{\text{GUE}}(\omega) - R_{\text{GOE}}(\omega)] \). The magnetoconductance is negative at \( \omega < \Delta \) and changes sign when the frequency is of the order of \( \Delta \), as depicted in Fig. 4. We believe that the state of the samples after illumination 1 correspond to this situation. This change of sign is however found to disappear for higher values of \( \gamma \), i.e., when the condition \( \delta G_{2} > |\delta G_{1}| \) is realized. We suggest that illuminations 3 and 4 correspond to this situation. The increase of \( \gamma \) with successive illuminations is necessary to explain our results. It cannot be due to the contribution of electron-electron interactions (whose contribution decreases with increasing electron density [23]) but possibly to the heightened losses in the etched GaAs substrate. We indeed observe a substantial decrease of \( Q \) after each illumination. The electromagnetic noise related to these losses is expected to contribute substantially to the level broadening [24]. Theory [6,7] also predicts, for the canonical ensemble, changes of sign of the magnetoconductance with temperature at \( T < \Delta \), which we have not observed, possibly due to the lack of experimental points in the regime where \( T \ll \Delta \).

In conclusion these results show that ac conductance measurements on isolated dots reveal new physical features. Giant magnetoconductance has been observed. Sign changes of the dissipative part are in agreement with the pioneering predictions of Gorkov and Eliashberg [22] on the sensitivity of the energy spectrum to time reversal breaking by a magnetic field. The reactive part, whose amplitude is of the same order of magnitude as the real one, is in principle related to the orbital magnetism in the dots. However its sign is not yet understood.

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