Magnetic-field asymmetry of mesoscopic *dc* rectification in Aharonov-Bohm rings

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Fundamental Casimir-Onsager symmetry rules for linear response do not apply to nonlinear transport. This motivates the investigation of nonlinear *dc* conductance of mesoscopic GaAs/GaAlAs rings in a two-wire configuration. The second-order current response to a potential bias is of particular interest. It is related to the sensitivity of conductance fluctuations to this bias and contains information on electron interactions not included in the linear response. In contrast with the linear response, which is a symmetric function of magnetic field, we find that this second-order response exhibits a field dependence, which contains an antisymmetric part. We analyze the flux periodic and aperiodic components of this asymmetry and find that they only depend on the conductance of the rings, which is varied by more than an order of magnitude. These results are in good agreement with recent theoretical predictions relating this asymmetric response to the electron interactions.

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I. INTRODUCTION

Magnetotransport properties of mesoscopic systems are sensitive to interferences between electronic waves and exhibit characteristic signatures of phase coherence.[1] Among them are universal conductance fluctuations (UCF) leading to reproducible sample specific magnetoresistance patterns, which in a ring geometry are modulated by the flux periodic Aharonov-Bohm (AB) oscillations. Besides these effects on the linear conductance, it has been shown that mesoscopic systems exhibit rectifying properties related to the absence of spatial inversion symmetry of the disorder or confining potential. This gives rise to a quadratic term in the *I*-*V* relation: $I=G_1V+G_2V$.[2] This nonlinearity was predicted theoretically[2] and observed experimentally more than 15 years ago.[3] It was understood as a direct consequence of the sensitivity of conductance fluctuations to the Fermi energy with a characteristic scale given by the Thouless energy $E_c=h/\tau_D$, where $\tau_D=L^2/D$ is the diffusion time across the sample of size L. More recently, it has been pointed out that nonlinear transport coefficients do not obey Casimir-Onsager symmetry rules[4] and may include, in a two-wire measurement, a component antisymmetric in magnetic field with a linear dependence at low field. On the experimental side, the existence of a term linear both in field and current was found in macroscopic helical structures and attributed to magnetic self-inductance effects.[5] It was subsequently observed in carbon nanotubes[6] and suggested to be related to their helical structure. More generally, this field asymmetry of nonlinear transport has been theoretically shown to be related to electron-electron interactions both in chaotic[7] and diffusive[8] systems. At the single impurity level, this effect can be simply viewed as the modification of the electron density $dn(\vec{r})$ around a scatterer in the presence of a current through the sample.[9,10] Due to Coulomb interactions, this results in a modification of the potential around the impurity by a component linear in current. In a phase-coherent sample, this bias induced change of disorder potential dU_{dis} modifies the conductance fluctuations, giving rise to the nonlinear conductance G_2 defined as $G(V)=G_1+G_2V+\cdots$. Just like the chemical potential measured in a multiprobe transport measurement,[1] dU_{dis} exhibits field-dependent fluctuations, which have both symmetric and antisymmetric parts, including a component linear in B at low field. As a result, G_2 has a symmetric component in B and an antisymmetric one, respectively, equal to $G_2^{S,AS}=[G_2(B)\pm G_2(-B)]/2$, which both vary on the flux scale Φ_c characteristic of conductance fluctuations. The antisymmetric component only exists in the presence of electron-electron interactions. The typical amplitudes of these components have been calculated in a two-dimensional (2D) system[7,8,11] and yield the following for weak interactions:

$$\delta G_2^S \sim \delta G_1\left(\frac{e}{E_c}\right), \quad \delta G_2^{AS} \sim \gamma_{int}\frac{\delta G_1}{g}f\left(\left|\frac{\Phi}{\Phi_c}\right|\right)\delta G_2^S, \quad (1)$$

where the function $f(x)$ is equal to x for $0<x\ll 1$ and to 1 for $x\gg 1$. $g=\langle G_1\rangle$ and $\delta G_1\simeq 1$ are the average conductance and the typical amplitude of G_1 fluctuations in units of e^2/h. In the weak interaction regime investigated in Ref. 8, $\gamma_{int}=2\nu dU_{dis}(\vec{r})/dn(\vec{r})\ll 1$, where ν is the density of states per unit surface. In the self-consistent treatment of Coulomb interaction,[11,12] the interaction constant is determined by the ratio of typical charging energy of the sample, $\sim e^2/C$, and mean level spacing Δ as $\gamma_{int}=1/(1+C\Delta/2e^2)$ [the limit $\gamma_{int}\ll 1$ of Eq. (1) corresponds to $dU/dn=e^2/2C$].[13] The $1/g$ factor in Eq. (1) indicates that the field asymmetry should be easily detectable in low conductance samples, where g does not exceed 10, but is not observable in metallic mesoscopic systems.[14] Carbon nanotubes[15] are, in principle, good candidates to investigate this physics, but it is delicate to distin-

guish the effects due to the tube helicity from mesoscopic ones. Semiconducting samples are well suited for such investigations, since they combine rather low conductance and large sensitivity to small fluxes.[16–18] We have measured the nonlinear conductance of two terminal GaAs/GaAlAs rings with only a few conducting channels. We find that G_2, like G_1, exhibits both AB oscillations and UCF conductance fluctuations. We show evidence of the existence of an asymmetry in magnetic field in G_2 from which we deduce the amplitude of the electron interactions.

II. EXPERIMENTAL RESULTS

The square rings investigated in this experiment were obtained by shallow etching through an aluminum mask of a 2D electron gas (2DEG) of density $n_e=3.8\times10^{15}$ m^{-2} at the interface of a GaAs/GaAlAs heterojunction with Si donors. Contrary to previous experiments,[15–18] there is no electrostatic gate on the samples.[19] Due to depletion effects, the real width of the rings is smaller than the etched one and is determined from magnetotransport data. We present data on two rings of circumference $L=4.8$ μm and respective widths $W=0.3$ and 0.45 ± 0.05 μm. The elastic mean free path l_e extracted from the conductance at 4 K varies between 1 and 2 μm, which is less than the value of the initial 2DEG and comparable with the side of the square ring. *In situ* modifications of the samples were obtained by short illuminations with an electroluminescent diode resulting in an increase of width and conductance of the rings. It was also possible to change the disorder configuration by applying current pulses in the 10–50 μA range, which decrease the average conductance of the samples. Therefore, with only two samples, we could investigate a conductance ranging from $g=1$ to 20. The measurements were conducted via filtered lines in a dilution refrigerator between 25 mK and 1.2 K. The samples were biased with ac current of frequency $\omega/2\pi\simeq30$ Hz in the few nanoampere range, and voltage was measured with a low noise amplifier followed by lock-in amplifiers detecting the first- and second-harmonic responses $V_1\cos\omega t$ and $V_2\cos2\omega t$. The amplitude of the ac modulation was chosen to maximize the second-harmonic signal in the regime where it is still quadratic with the current modulation amplitude. In this regime, the second-order conductance G_2 is simply related to V_2 and V_1 by $G_2=-2V_2I/V_1^3$. As shown in Fig. 1, both $G_1(B)$ and $G_2(B)$ exhibit h/e periodic AB oscillations modulated by UCF fluctuations.

More remarkable, whereas $G_1(B)$ is a symmetric function of magnetic field as expected in a two-wire configuration, $G_2(B)$ exhibits a component antisymmetric in field G_2^{AS}. To compare the field asymmetry of G_2 on the various samples (Fig. 2), we extract the amplitude of UCF and AB oscillations from the Fourier transform of $G_2^{S,AS}(B)$ and $G_1(B)$. The integrated UCF and AB peaks are noted as $\delta G_2^{S,AS}$ and $\delta\tilde{G}_2^{S,AS}$ and similarly for G_1. These averages performed on a flux range much larger than Φ_c do not depend on this range. We find that all these quantities are aligned on logarithmic plots as a function of g. We first note the relatively large amplitudes of the AB oscillations $\delta\tilde{G}_2^{S,AS}$, which are of the order of

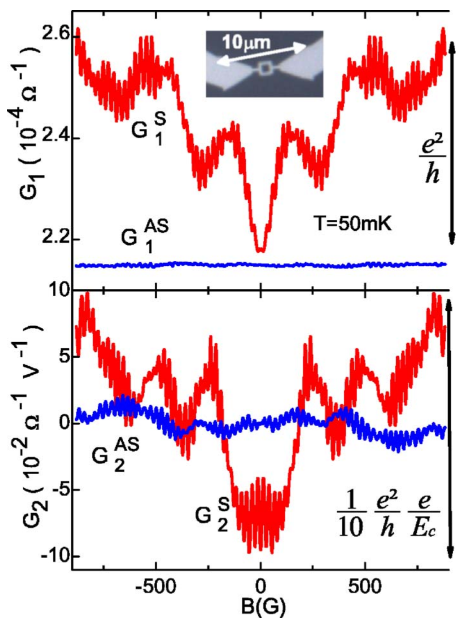

FIG. 1. (Color online) Field dependence of $G_1^{S,AS}$ and $G_2^{S,AS}$ (G_1^{AS} shifted). These data were obtained on ring 1 in its initial state for a current of 10 nA. Inset: micrograph of ring 1.

the UCF components $\delta G_2^{S,AS}$ in contrast with the related quantities in G_1.[20] The decrease of δG_1 and $\delta\tilde{G}_1$ to values much below 1 at low g seems to be at odds with the universal character of conductance fluctuations in G_1 established deep in the diffusive regime $g\gg1$. However, this universal regime is not expected for $g\simeq1$. The even larger dependence measured in $\delta G_2^S(g)$ indicates, according to Eq. (1), that $E_c=g\Delta$ decreases with g. This finding can be attributed to the physics involved in the illumination process, which increases the width of the rings (resulting in a decrease of Δ) and also decreases the elastic scattering time. Finally, we find that the

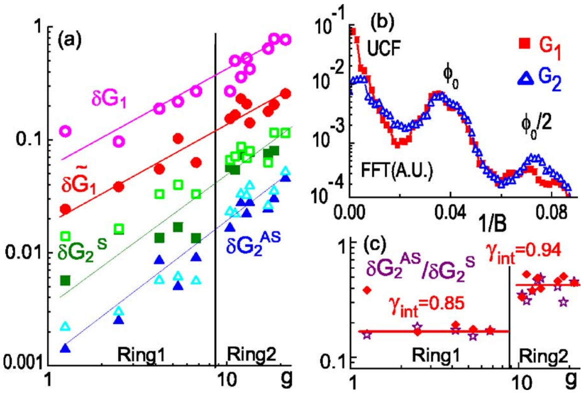

FIG. 2. (Color online) (a) Amplitudes of the UCF and AB components of G_1 in units of e^2/h, and G_2^S and G_2^{AS} in $(\Omega\,\mathrm{V})^{-1}$ as a function of the conductance of the rings. The open and closed symbols correspond to UCF and AB integrated peaks (straight lines are guides to the eyes). (b) Fourier transforms of G_2 and G_1 renormalized so that the amplitude of the AB peaks are identical. (c) Conductance dependence of the ratio $\delta G_2^{AS}/\delta G_2^S$ for UCF (open stars) in comparison with Eq. (2) (diamonds), where $\gamma_{int}=0.85$ for ring 1 and 0.94 for ring 2.

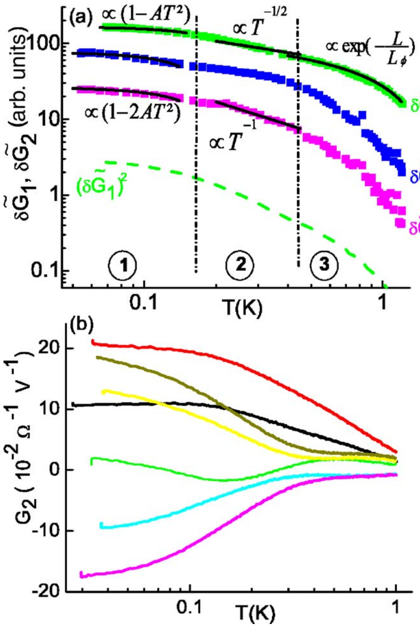

FIG. 3. (Color online) (a) Temperature dependence of the amplitude of the AB oscillations in G_1, G_2^S, and G_2^{AS} (ring 2). Continuous lines are the fits corresponding to the regions (1) $k_BT \ll E_c$ and (2) $k_BT \gg E_c$ but $L \ll L_\phi$ (3) $L > L_\phi$. (b) Examples of temperature dependence of G_2 in ring 2 for different values of the magnetic field between −1000 and 1000 G.

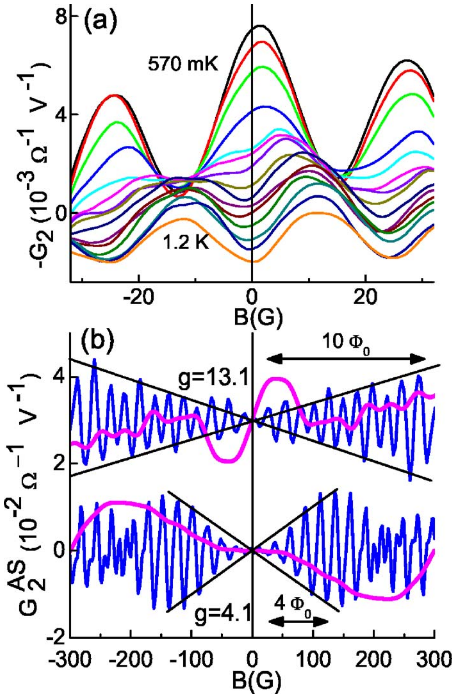

FIG. 4. (Color online) (a) Low-field dependence of G_2 on ring 1 for different temperatures. (b) Field dependence of G_2^{AS} after low-pass (light line) and high-pass (bold line) filtering on ring 1 and ring 2, respectively, lower and upper curves (shifted for clarity). Note the extinction of AB oscillations in the vicinity of $B=0$.

ratio $\delta G_2^{AS}/\delta G_2^S$ depends only slightly on g and is equal to 0.3 ± 0.2 [see Fig. 2(c)]. This result is *a priori* in contradiction with Eq. (1) from which a $1/g$ dependence of $\delta G_2^{AS}/\delta G_2^S$ is expected. However, Eq. (1) is only valid for $\gamma_{int} \ll 1$ in partially closed quantum dots, i.e., whose classical resistance is dominated by the contacts but still much lower than quantum resistance. It was recently found by one of us[21] using random matrix theory that in these systems, δG_2^S also strongly depends on γ_{int} when it is not negligible compared to 1, and in the limit $\Phi \gg \Phi_c$,

$$\delta G_2^{AS}/\delta G_2^S = 1/\sqrt{1+2(g/\delta G_1)^2(1/\gamma_{int}-1)^2}, \qquad (2)$$

identical to Eq. (1) in the limit of small γ_{int}. It is interesting to note that Eq. (2) predicts $\delta G_2^{AS}/\delta G_2^S$ independent of g in the limit $\gamma_{int}=1$. It describes our experimental results remarkably well, yielding $\gamma_{int} \simeq 0.94\pm0.02$ for ring 2 and $\gamma_{int} \simeq 0.85\pm0.02$ for ring 1. These values are very close to the estimated values of $\gamma_{int}=1/(1+C\Delta/2e^2) \simeq 0.98$ from the geometry of our samples. Our determination of the interaction constant from the dimensionless quantity $\delta G_2^{AS}/\delta G_2^S$ is much more accurate than the estimation done in Ref. 17 from the analysis of the low-field component of G_2^{AS} extracted from differential conductance measurements, but consistent with it. However, it seems that no symmetric component in G_2 could be detected in this work. Finally, our results show that our rings are in a highly interacting regime and behave as partially closed quantum dots due to their relatively long mean free path.

The temperature dependence of $\delta\tilde{G}_1$ and $\delta\tilde{G}_2^{S,AS}$ are shown on Fig. 3(a). As already observed,[22] $\delta\tilde{G}_1$ is only weakly T dependent below the Thouless energy such as $1-AT^2$, with $A=2(k_B/E_c)^2$, and decays at higher temperature as $T^{-1/2}$. Deviations above 0.5 K are consistent with $\exp(-L/L_\phi(T))$, with a T dependence of the phase-coherence length as $T^{-1/2}$, but this last fit on a small range of temperature is not unique and just indicative. $\delta\tilde{G}_2^{S,AS}$ have similar T dependences, which are nearly identical to $[\delta\tilde{G}_1(T)]^2$ with a T^{-1} decay in the limit $k_BT \gg E_c$ with $L \ll L_\phi$, in agreement with theoretical predictions.[8,11] The temperature dependence of G_2 at fixed magnetic fields is depicted in Fig. 3(b). In the same way as observed on G_1,[23] $G_2(T)$ exhibits a nonmonotonous variation with temperature on the scale E_c, which randomly fluctuates with magnetic field. In some cases, we also observed [see Fig. 4(a)] that the phase of the AB oscillations in G_2 depends on temperature. Surprisingly, it remains pinned either to 0 or π at zero field with the appearance of a second-harmonic contribution in the region of temperature where the sign change occurs. At larger fields, the phase takes any value between 0 and π. Note that this last effect is observed both in G_1 and G_2 as a result of phase modulation of the AB oscillations by the UCF, but these phase modulations are symmetric in field on G_1 and not on G_2. In short, we find that AB oscillations on G_2 are symmetric in zero field and the asymmetry only appears at higher fields. This is also clearly seen in the AB oscillations on G_2^{AS}. We observe in all samples that their amplitudes vanish linearly at zero field [Fig. 4(b)].

III. MESOSCOPIC RECTIFICATION FROM SEMICLASSICS

We now propose a simple explanation of the extinction of AB oscillations at low fields in G_2^{AS} linked to the larger AB component observed in G_2 compared to G_1. In the semiclassical approximation ($k_f l_e \gg 1$),[24] it is possible to express the conductance of a phase-coherent ring in terms of interference between scattering amplitudes of electronic waves. $G_Q = \mathrm{Re}[\Sigma_{i,j} A_i A_j \exp i\phi_{ij}(B)]$, where indices i and j run on all pairs of possible diffusive trajectories going from one terminal to the other and $\phi_{ij}(B) = \phi_i - \phi_j + 2\pi B S_{ij}/\Phi_0$. The phase at zero field ϕ_i is the integral $(1/\hbar)\int_i U_{dis}(r(\vec{t}))dt$ on the diffusive trajectory i assuming that the screened disordered potential U_{dis} varies smoothly on the electron Fermi wavelength, which is a reasonable assumption in a 2DEG. S_{ij} is the surface comprised between trajectories i and j. Onsager symmetry rules imply that the two terminal magnetoconductance takes the following form at zero temperature:

$$G_Q = \sum_{i,j} A_i A_j \cos(\phi_{ij})\cos\left(\frac{2\pi B S_{ij}}{\Phi_0}\right). \quad (3)$$

In the presence of a current through the sample associated with a potential drop V, the local electronic density and, consequently, the scattering potential are modified with a term $dU_{dis}(\vec{r},V)$, which induces phase shifts, $d\phi_{ij} = \int_i - \int_j (1/\hbar) dU_{dis}(r(\vec{t}))dt$. The quantity $G_2 = [dG(V)/dV]_{V=0}$ is then directly related to these phase shifts through

$$G_2 = \sum_{i,j} A_i A_j \left(\frac{d\phi_{ij}}{dV}\right)\sin(\phi_{ij})\cos\left(\frac{2\pi B S_{ij}}{\Phi_0}\right). \quad (4)$$

Since $d\phi_{ij}$ increases with the length of the interfering trajectories i and j, long trajectories encircling the ring (AB oscillations) contribute more than the trajectories within the same branch of the ring (UCF). This explains the larger relative amplitude of the AB oscillations and the larger harmonic content in G_2 compared to G_1 [see Figs. 2(a) and 2(b)]. A similar behavior was also observed in rings interrupted by tunnel junctions.[20] In a diffusive open system, dU_{dis} is of the order of eV and the main contribution to the typical value of $d\phi_{ij}$ is $eV\tau_D/\hbar$, is independent of B, which leads to the expression of δG_2^S of Eq. (1). This contribution is strongly attenuated for a ballistic system with strong interactions such as in Ref. 11, where most of the potential drop takes place at the contacts. Other contributions to δG_2 of the order of $1/g$ are obtained by taking into account mesoscopic nonlocal field-dependent fluctuations in the potential $dU_{dis}(\vec{r},B)$, with symmetric and antisymmetric components in B, both of the order of $\gamma_{int} eV \delta G_1/g$ at high flux compared to Φ_c. The antisymmetric component in $dU_{dis}(\vec{r},B)$ is expected to typically vary like $\gamma_{int}(\delta G_1/g)V\Phi/\Phi_c$ at low flux, also with AB oscillations in $\sin(2\pi\Phi/\Phi_0)$ of smaller relative amplitude $\delta\tilde{G}_1/\delta G_1$. We can then deduce from Eq. (4) the main contributions to the low-field AB oscillating part of $\tilde{G}_2^{AS}$, which originates from pairs of trajectories encircling the rings:

$$\tilde{G}_2^{AS} = \frac{\delta G_1}{g\hbar}\gamma_{int}\sum_{i,j} eA_iA_j\tau_{ij}\sin(\phi_{ij})\frac{\Phi}{\Phi_c}\cos\left(\frac{2\pi\Phi}{\Phi_0}\right). \quad (5)$$

There is also a smaller contribution of terms in $\sin(2\pi\Phi/\Phi_0)$ of the order of $\delta\tilde{G}_1/\delta G_1$, which is less than 0.2 in our experiments. This provides an explanation for the linear increase of the amplitude of the AB oscillations of G_2^{AS} in a flux range Φ_c for rings with $g > 1$, as shown in Fig. 4(b). However, this effect may not exist in very narrow rings,[18] where conductance fluctuations are not observed. It is interesting that this simple heuristic model can also explain the larger value of G_2^S in a diffusive compared to a ballistic system and the different values of the ratio G_2^{AS}/G_2^S of the orders of γ_{int}/g and 1 in a diffusive and a ballistic system, respectively.

IV. CONCLUSION

In conclusion, we have shown evidence of a field asymmetry on the second-order response of GaAs/GaAlAs rings of mesoscopic origin, which contains both AB oscillations and conductance fluctuations. This asymmetry is characterized by $\delta G_2^{AS}/\delta G_2^S$ and analyzed within theoretical predictions expressing this ratio with only two parameters, namely, the dimensionless conductance of the rings and the interaction constant whose value can be determined as $\gamma_{int} = 0.90 \pm 0.05$. We have also found that the relative amplitude of the AB oscillations compared to the UCF is much larger in G_2 than in G_1, with the existence of a linear low-field modulation in the AB oscillations in the antisymmetric component of G_2. These effects can be understood within a simple semiclassical description of quantum interference. Recently, it has come to our attention that related experimental work on gated quantum dots[17] and small Aharonov-Bohm rings[18] have appeared.

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[1] See, e.g., *Mesoscopic Phenomena in Solids*, edited by B. L. Altshuler, P. A. Lee, and R. A. Webb (Elsevier, Amsterdam, 1991); Y. Imry, *Introduction to Mesoscopic Physics* (Oxford Universty Press, New York, 1997).

[2] B. L. Altshuler and D. E. Khmel'nitskii, JETP Lett. **42**, 359 (1985); A. I. Larkin and D. E. Khmel'nitskii, JETP **64**, 1075 (1986).

[3] R. A. Webb, S. Washburn, and C. P. Umbach, Phys. Rev. B **37**, 8455 (1988); P. G. N. de Vegvar, G. Timp, P. M. Mankiewich, J. E. Cunningham, R. Behringer, and R. E. Howard, Phys. Rev. B

38, 4326 (1988).
[4] L. Onsager, Phys. Rev. **38**, 2265 (1931); H. G. B. Casimir, Rev. Mod. Phys. **17**, 343 (1945).
[5] G. L. J. A. Rikken, J. Fölling, and P. Wyder, Phys. Rev. Lett. **87**, 236602 (2001).
[6] V. Krstic *et al.*, J. Chem. Phys. **117**, 11315 (2002).
[7] D. Sanchez and M. Büttiker, Phys. Rev. Lett. **93**, 106802 (2004); Int. J. Quantum Chem. **105**, 906 (2005).
[8] B. Spivak and A. Zyuzin, Phys. Rev. Lett. **93**, 226801 (2004); E. Deyo, B. Spivak, and A. Zyuzin, Phys. Rev. B **74**, 104205 (2006).
[9] R. Landauer, Z. Phys. B **21**, 247 (1975).
[10] W. Zwerger, L. Bönig, and K. Schönhammer, Phys. Rev. B **43**, 6434 (1991).
[11] M. L. Polianski and M. Büttiker, Phys. Rev. Lett. **96**, 156804 (2006).
[12] T. Christen and M. Büttiker, Europhys. Lett. **35**, 523 (1996).
[13] We note that for a metallic sample, Δ is very small and γ_{int} is very close to 1.
[14] R. Haussler, E. Scheer, H. B. Weber, and H. V. Lohneysen, Phys. Rev. B **64**, 085404 (2001); C. Terrier *et al.*, Europhys. Lett. **59**, 437 (2002).
[15] J. Wei, M. Shimogana, Z. Wang, I. Radu, R. Dormaier, and D. H. Cobdem, Phys. Rev. Lett. **95**, 256601 (2005).
[16] A. Löfgren, C. A. Marlow, I. Shorubalko, R. P. Taylor, P. Omling, L. Samnelson, and H. Linke, Phys. Rev. Lett. **92**, 046803 (2004).
[17] D. M. Zumbühl, C. M. Marcus, M. P. Hansan, and A. C. Gossard, Phys. Rev. Lett. **96**, 206802 (2006).
[18] R. Leturcq, D. Sanchez, G. Gotz, T. Ihn, K. Ensslin, D. C. Driscoll, and A. C. Gossard, Phys. Rev. Lett. **96**, 126801 (2006).
[19] As shown in G. L. J. A. Rikken and P. Wyder, Phys. Rev. Lett. **94**, 016601 (2005), a gate potential contributes to a term linear in H in G_2 of purely classical origin.
[20] A. Van Oudenaarden, Yu. N. Nazarov, and J. E. Mooij, Phys. Rev. B **57**, 8816 (1988).
[21] M. L. Polianski and M. Büttiker, cond-mat/0701024.
[22] S. Washburn and R. A. Webb, Adv. Phys. **35**, 375 (1986).
[23] B. Spivak, A. Zyuzin, and D. H. Cobden, Phys. Rev. Lett. **95**, 226804 (2005).
[24] K. Richter, D. Ullmo, and R. A. Jalabert, Phys. Rev. B **54**, R5219 (1996).