Coulomb blockade related to a localization effect in a single tunnel-junction/carbon-nanotube system

Junji Haruyama, Izumi Takesue, Tetsuro Hasegawa, and Yuki Sato
Aoyama Gkuin University, Department Electrical Engineering and Electronics 6-16-1 Chitosedai, Setagaya, Tokyo 157-8572 Japan
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We report on Coulomb blockade caused by the high impedance external electromagnetic environment (EME) related to a localization effect in a single tunnel-junction/carbon-nanotube system. Observed Coulomb blockade, supported by a linear temperature dependence of zero-bias conductance, mathematically follows phase correlation theory, which explains the roles of EME and implies that tunneling of electrons is suppressed by transferring the energy to its EME with a total impedance $Z_t(\omega)$ higher than quantum resistance. Our high $Z_t(\omega)$, however, is strongly associated with the antilocalization effect without any energy dissipation but actually contributes to Coulomb blockade, because its phase modulation by magnetic field modulates also Coulomb blockade. Is the energy transfer to such high impedance EME actually indispensable for Coulomb blockade?

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Coulomb blockade, a typical phenomenon associated with single electron tunneling, has been successfully observed in a variety of systems. Recently, its correlation with a phase of electron waves and spins in the external electromagnetic environment (EME) has attracted much attention because Coulomb blockade is very sensitive to its EME. Most of such studies have been performed in multijunction systems. On the contrary, no work has reported it in single-junction systems. It is because Coulomb blockade in single-junction systems is too sensitive to the electron phase in its EME and hence provides characteristics quite different from those in multijunctions. It has been well known as phase correlation (PC) theory.[1,2]

PC theory explains the roles of the phase fluctuation of EME on Coulomb blockade in single-junction systems, from the following two viewpoints: (1). The tunneling of electrons is suppressed by transferring the energy to the EME by exciting environmental modes (e.g., *LC* and *RC* circuit modes), leading to Coulomb blockade. It is based on the quantum-mechanical treatment of the total impedance of EME $Z_t(\omega)$ replaced by a set of harmonic oscillators (i.e., a set of *LC* circuits in a circuit model) with energy quantum $\hbar\omega$, in accordance with the spirit of Caldeira and Leggett.[7] (2). Phase fluctuation of the EME fluctuates the junction surface charges (e.g., by coupling with the zero-point oscillation or through a commutation relation between phase fluctuation $\tilde{\varphi}$ in the environment and charge fluctuation $\tilde{Q}$ on the junction surface (i.e., $[\tilde{\varphi},\tilde{Q}]=ie$), smearing out Coulomb blockade. To realize the first interpretation and avoid the second interpretation, the real part of $Z_f(\omega)$ (Re$[Z_f(\omega)]$ must be much larger than resistance quantum ($R_Q=h/e^2\sim 25.8$ KΩ). This is the key factor in PC theory. $Z_t(\omega)$ also must be closely connected to a single junction as a high impedance transmission line R_L ($\gg R_Q$), to avoid the second interpretation.

To our best knowledge, an interpretation has been theoretically studied well,[1–3] whereas only a few works experimentally reported it only by the data fitting to conductance vs voltage features [i.e., data fitting by Eqs. (1)–(3) shown in later section].[4–6] None directly confirmed *how the energy transfer was performed in actual systems*, nevertheless, it is a very important problem from the viewpoint of quantum mechanics with energy dissipation.

On the contrary, the second interpretation has been experimentally well confirmed (e.g., by Cleland[8] and Delsing,[9] carefully treating the contribution of parasitic capacitance). Cleland *et al.* also explained the Coulomb blockade by the junction charge fluctuation calculated from a quantum Langevin equation employing Nyquist voltage noise caused in the *LCR* transmission line. It should be noticed that his model did not employ any energy dissipation in the EME. We therefore ask a question ''*Is the first interpretation (energy transfer) actually indispensable to yield Coulomb blockade in realistic systems*?'' The purpose of this work is to clarify that question.

Here, since the connection of the high impedance lead line R_L automatically leads to a high $Z_t(\omega)$, it is difficult to distinguish the first interpretation from the second one. In this report, we try to utilize a localization effect, a typical phase interference effect of electron waves, as the high $Z_t(\omega)$. From the viewpoint of the first interpretation, $Z_t(\omega)$ in the *LC* mode should correspond to a delay of surface electron charge propagation related to electron-phonon scattering in the EME in actual systems. In contrast, when a single junction is coupled only with a resistive wire (an Ohmic resistor) described by the frequency independent impedance $Z(\omega)=R$, the system is *RC* circuit. Energy transfer by exciting this *RC* mode should directly coincide with electron-phonon scattering in the Ohmic resistance in actual systems. Here, since a localization effect is basically an elastic process, any energy cannot be transferred there in accordance with Landauer theory.[11] If, therefore, Coulomb blockade is observable even in this system, it will imply that the first interpretation (energy transfer) is not necessarily required in actual systems. In other words, only avoiding the phase fluctuation by connecting a high R_L can yield Coulomb blockade. In this paper, we used multiwalled carbon nanotubes (MWNT's) to introduce a localization effect because it is already known that it exhibits weak localization with resistance higher than R_Q,[10] nevertheless, weak localization is a small quantum correction in the theory of condensed matters.[11]

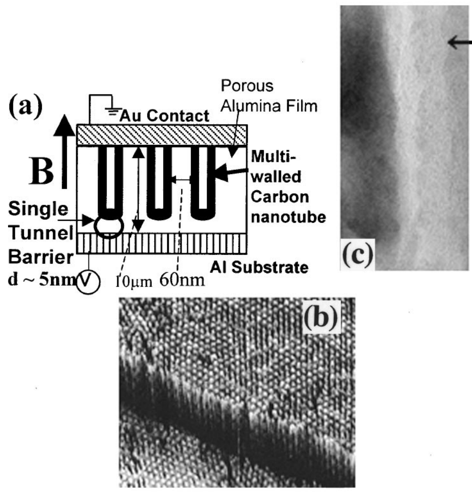

FIG. 1. (a) Schematic cross section of the sample, an array of single tunnel junctions connected to multi-walled carbon nanotubes (MWNT's) (i.e., array of $Al/Al_2O_3/MWNTs$). We have simply interpreted the measurement result as a superposition of each MWNT's characteristic, because of the large enough spacing among the MWNTs and the very high uniformity of structure parameters (e.g., half width of pore diameter distribution less than 10%). (b) Scanning electron microscope (SEM) image of the exposed MWNT array. (c) Cross sectional high resolution transmission electron microscope (CSHRTEM) image of one-side shells of MWNT with about 26 layers, as indicated by the arrow.

Although Coulomb oscillation also has been already reported in single-walled carbon nanotube systems with multi-tunnel junctions,[12] it should be emphasized that this report has quite a different physical meaning from those.

We measured the static electric characteristics of the sample shown in Fig. 1(a). Figure 2(a) clearly exhibits a zero-bias conductance (G_0) anomaly. The shape of G_0 anomaly is drastically varied near $T=5$ K and the shape at 2 K is quite different from the inset.[13,14] It is evidence that the nanomaterials connected to single tunnel junctions strongly contributes to the G_0 anomaly. The G_0 anomaly in Fig. 2(a) can be mathematically fit by phase correlation (PC) theory as shown in Fig. 2(b) and as explained in the later paragraph, whereas that in the inset of Fig. 2(a) cannot be directly fit by PC theory. Its first derivative (i.e., dG/dV vs V curve) is fit by Nazarov's theory introducing an electron-electron interaction to the EME of Coulomb blockade[13] as shown in the left inset of Fig. 2(b). This large difference is also evidence that Coulomb blockade in the single-junction system can be much influenced by mesoscopic phenomena in the nanowires directly connected.

Figure 2(c) distinguishes the temperatures to the following three regions, (1). *Above 10 K:* linear G_0 vs log(T) relation, (2) *5–10 K:* its saturation region, and (3) *Below 5 K:* linear G_0 vs temperature relation (see the upper inset). This linear G_0 vs temperature relation was also observable in the lower inset. This provides strong qualitative and quantitative evidence of Coulomb blockade in an array of single junctions located in parallel (i.e., as a temperature dependence of averaged G_0) as we have implied in Ref. 13, when one neglects the influence of the external environment. In addition, this linear relation disappears in the sample without a tunnel barrier. It also supports the presence of Coulomb blockade. The temperature of 5 K also agrees with that at which the shape of the G_0 anomaly starts to change in Fig. 2(a). It is also consistent with this temperature region.

In order to clarify the correlation of this Coulomb blockade with PC theory, we first numerically calculate a G vs V curve, normalized by tunneling resistance (R_t) and the number of junctions, using Eqs. (1)–(3) from PC theory.[1,2] We then fit the G vs V curve measured at 2 K in the Coulomb blockade regime in Fig. 2(a) by the calculation result. Here,

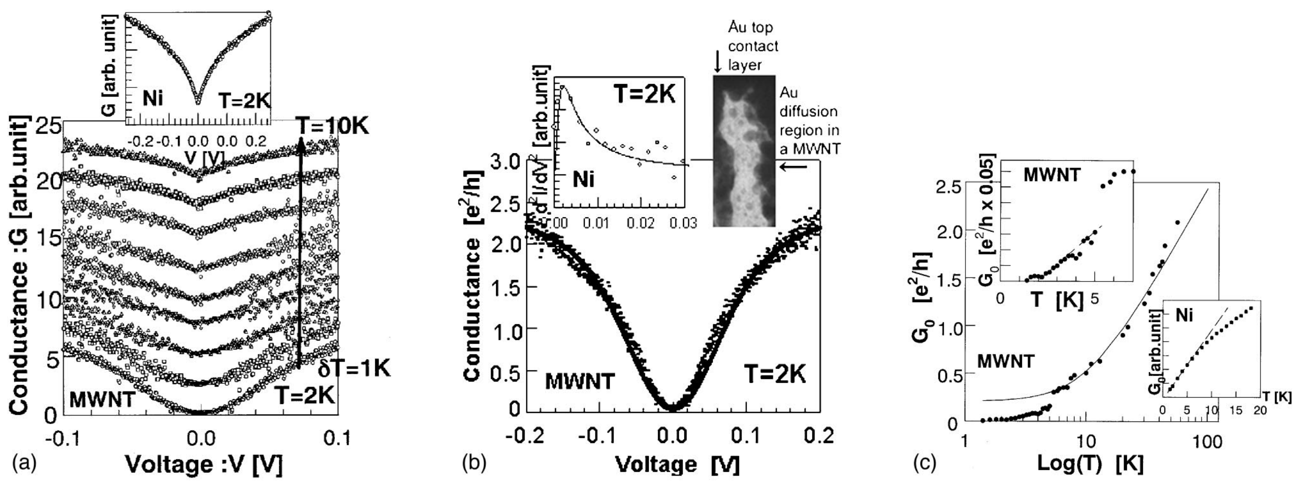

FIG. 2. (a) $Al/Al_2O_3/MWNT$ array: Temperature dependence of a typical conductance ($G=dI/dV$) vs voltage (V) curve. Inset: G vs V curve of an Al/Al_2O_3/Ni-nanowires array, fabricated in the porous Alumina membrane with the exactly same structure parameters as Fig. 1(a) (Ref. 13). (b) Data fitting to G vs V curve of (a) by phase correlation theory. The solid line was numerically calculated from Eqs. (1)–(3). Left inset: Data fitting to dG/dV vs V curve of the (a) inset by Nazarov's theory (Ref. 13). Right inset: CSHRTEM image of the top part of MWNT. (c) Temperature (T) dependence of G_0 shown in (a). The solid line is the result calculated by Eq. (4). Upper inset: Linear G_0 vs temperature relation at temperatures below 5 K in (c). Lower inset: Ni-nanowire system; Temperature dependence of G_0, indicating a linear G_0 vs temperature relation at low temperatures.[13]

R_t is 300 KΩ ($\gg R_Q$) from the measurement and we also employed a lumped RC as the simplest case, because the inductance (L) related characteristics of MWNT is not yet clarified. Hence, the fitting parameters are the resistance of external environment ($R_{\rm ext}$) and junction capacitance (C) as $R_Q/R_{\rm ext}$ and $\hbar\omega_{RC}/kT$, where $\omega_{RC}=1/(R_{\rm ext}C)$.

$$I(V)=\frac{1-e^{-\beta \mathrm{eV}}}{eR_1}\int_{-\infty}^{+\infty}dE\,\frac{E}{1-e^{-\beta E}}P(\mathrm{eV}-E), \qquad (1)$$

$$P(E)=\frac{1}{2\pi\hbar}\int_{-\infty}^{\infty}dt e^{J(t)+i(E/\hbar)t}, \qquad (2)$$

$$J(t)=2\int_{-\infty}^{\infty}\frac{d\omega}{\omega}\frac{\mathrm{Re}[Z_t(\omega)]}{R_Q}\frac{e^{-i\omega t}}{1-e^{-\beta\hbar\omega}}, \qquad (3)$$

where β is the $1/kT$ and $Z_t(\omega)=1/[i\omega C+Z(\omega)^{-1}]$ is the total EME impedance consisting of junction capacitance C in parallel with an external environment impedance $Z(\omega)=R_{\rm ext}$ in a RC circuit model. $J(t)$, $P(E)$, and $I(V)$ are phase correlation functions, Fourier transform of $J(t)$, and the tunnel current, respectively.

These three equations well represent the general argument of PC theory mentioned in the introduction part. Phase fluctuation of the junction surface charges caused by the tunneling electrons and its time evolution lead to Eq. (3) through the commutation relation between charge and phase. Tunneling current (probability) is obtained from Eq. (1) by perturbatively treating the tunneling Hamiltonian using Fermi's golden rule and $P(E)$, which is interpreted as a probability density for the tunneling electrons to transfer the energy by exciting the EME mode described by $J(t)$. If $\mathrm{Re}[Z_t(\omega)]/R_Q$ is much smaller than one in Eq. (3), $P(E)$ becomes delta function $\delta(E)$ and, thus, Coulomb blockade disappears in Eq. (1) (i.e., the first interpretation). In that case, $J(t)$ and then phase fluctuation $\tilde{\varphi}$ can also be neglected. Hence, charge fluctuation $\tilde{Q}$ diverges due to $[\tilde{\varphi},\tilde{Q}]=ie$, smearing Coulomb blockade voltage $e/2C$ (i.e., the second interpretation).

As shown by the solid line in Fig. 2(b), the measurement and calculation results are in excellent agreement in our weak tunneling case (i.e., R_t of 300 K$\Omega > R_Q$). The best fitting gives the $R_{\rm ext}$ of 450 KΩ and $\mathrm{Re}[Z_t(\omega)]$ with the same order value as $R_{\rm ext}$, which implies the value larger than R_Q. This $R_{\rm ext}$ of 450 KΩ should be due mainly to the resistance of MWNT ($R_{\rm NT}$) in our system, because only MWNT was directly connected to the single junction and the resistance of the gold contact layer with the gold/MWNT interface was at most on the order of 100 Ω. The low interface resistance originates from the diffusion of gold particles deposited as the top contact layer into the MWNT by high-temperature annealing, as shown in the right inset of Fig. 2(b). The value of 450 KΩ as the resistance of MWNT is also in good agreement with that in a previous report.[10] We, therefore, conclude that the MWNT acts as a high impedance EME for this Coulomb blockade.

The origin of this high impedance of MWNT is the key point for this report. As we expected, it can be qualitatively understood as a result of a localization effect from the curve fitting shown in Fig. 2(c). As shown by the solid line, the G_0 vs temperature characteristic is in nice agreement with the following formula of two-dimensional (2D) weak localization of MWNT,[10] except for the Coulomb blockade temperature region:

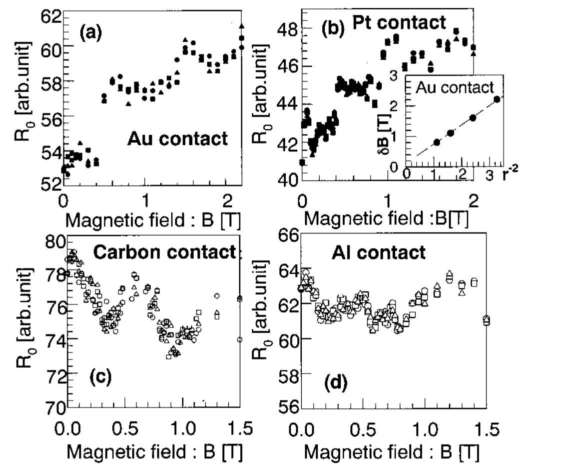

FIG. 3. (a) Magnetoresistance (MR) in the Coulomb blockade temperature regime in the Gold contact sample. Magnetic field was applied along the tube axis as shown in Fig. 1(a). (b) MR oscillation in the Platinum contact sample. (c) MR oscillation in the Carbon contact sample. (d) MR oscillation in the Aluminum contact sample. Inset of (b): Observed MR oscillation period δB vs inverse of square mean radius (r^{-2}) [$10^{-3}\times\mathrm{nm}^{-2}$] of MWNT obtained from HRTEM image in each sample. The dot line means a linear relation, supporting AAS oscillation.

$$G(T)=G(0)+\frac{e^2}{2\pi^2\hbar}\frac{n\pi d}{L}\ln\left[1+\left(\frac{T}{T_c(B,\tau_s)}\right)^p\right], \qquad (4)$$

where n, d, L, and τ_s are the number of shells, the diameter of the inner shell of the MWNT, the length of the MWNT, and the relaxation time of spin-flip scattering, respectively. The best fitting gives $n=18$, $p=2.1$, and $T_c=10\,\mathrm{K}$. Here, the Coulomb blockade obstructs the observation of the temperature dependence of the localization effect at the low temperatures. It is, however, revealed by applying magnetic field along the tube axis as shown in Fig. 3 and as explained in the Altshuler-Aronov-Spivak (AAS) effect the latter part. The positive magnetoresistances (MRs) around zero magnetic field in Figs. 3(a) and 3(b) imply that this localization is an antilocalization effect, in which phases of electron waves are locked in opposite to that of weak localization (i.e., with the phase difference of π). It has been already reported in a two-dimensional gold film[16] and the AAS effect in magnesium tube,[15] with strong spin-orbit interaction. Since the gold particles diffuse into the MWNT in our structure as shown in the right inset of Fig. 2(b), it can cause this antilocalization qualitatively similar to Refs. 15 and 16. In contrast, the samples with the top contact layers of carbon and aluminum, which have less spin-orbit interaction, exhibit negative MRs around $B=0$, which are consistent with weak localization, as shown in Fig. 3(c) and 3(d), respectively. This is strong evidence that the resistance of the MWNT is associated with a localization effect.

Consequently, these results and analyses [i.e., (1) the linear G_0 vs temperature dependence supporting the presence of the Coulomb blockade, (2) Coulomb blockade is fit by Eqs. (1)–(3) with about 450 KΩ of the EME impedance, (3) the EME impedance can exist only in the MWNT directly connected to the junction, and (4) the origin of the highly resistive MWNT is associated with antilocalization effect] implies that the localization effect in the MWNT yields $\mathrm{Re}[Z_t(\omega)]$ higher than R_Q by coupling a junction capacitance C as the *RC* mode in PC theory and contributes to Coulomb blockade.

Since, however, antilocalization effect is basically an elastic process, the tunneling electrons cannot transfer any charging energy there as discussed in the introduction. Therefore, we conclude that (1) the energy transfer in the EME with an impedance higher than R_Q is not necessarily required and (2) Coulomb blockade in single-junction systems can be caused only by avoiding the phase fluctuation by connecting a high $R_L(\gg R_Q)$. Of course, tunneling of electrons must be suppressed by transferring the energy somewhere from the point of view of quantum mechanics with energy dissipation, leading to Coulomb blockade. It will be performed in the other part of the external environment (i.e., metal reservoirs in which Landauer theory assumes quick energy dissipation) with an impedance smaller than R_Q.

In accordance with this discussion, $P(E)$ in Eqs. (1) and (2) should be reinterpreted as *the other probability*, which is not associated with the energy transfer probability of the tunneling electrons to the EME. Here, $P(E)$ is the Fourier transform of $J(T)=\langle[\tilde{\varphi}(t)-\tilde{\varphi}(0)]\tilde{\varphi}(t)\rangle$, a time evolution of phase fluctuation $\tilde{\varphi}$ in the EME, and the origin of the phase was defined as $\varphi(t)=e/\hbar\int_{-\infty}^{t}dtV(t)$, where $V(t)=Q(t)/C_j$ is the voltage across the tunnel junction.[1] The phase interference effect in localization also originates from this definition. Hence, $J(t)$ is a time evolution of $\tilde{\varphi}$ but should be attached to the localization effect so as not to destroy phase coherence in the MWNT. In the sense, $P(E)$ may be reinterpreted as a transmission probability of electrons, associated with $J(t)$ in the localization regime, in the MWNT.

Otherwise, we may have to perform more careful data fitting from the following points. (1) Junction capacitance C: We used C obtained in Ref. 13. Since the C was estimated from data fitting by Nazarov's theory, it is not yet experimentally confirmed, (2) Parasitic capacitance C_p: We did not take into consideration the influence of the C_p of MWNT. When we defined $L=\tau\times c$ (where $\tau\sim h/\mathrm{eV}$, c is the velocity of light in vacuum) as the geometry for an effective C_p based on the horizontal model[9,17] and included the C_p in the data fitting, it did not exhibit perfect agreement. C_p for better agreement should be smaller than that calculated from $L=\tau\times c$. To explain this difference, a smaller velocity instead of "c" may have to be employed, because our MWNT has a very disordered surface, and (3) *LCR* model: We also have employed the *RC* mode as a lumped circuit model in PC theory here. Since, however, MWT has distributed L, R, and C including this C_p in the actual system, the *LCR* transmission line model will have to be introduced.[1] However, even if apart from these data fitting problems, the linear G_0 vs. temperature dependence (the high impedance EME, which exists only in the MWNT directly connected to the junction) and its dependence on the localization effect will support our conclusion.

In order to reconfirm that the localization effect actually contributes to Coulomb blockade, we modulate the phase of electron waves in the MWNT by applying a magnetic field in the Coulomb blockade temperature regime. Since the Coulomb blockade is basically independent of the magnetic field applied, this result will clarify it. As shown in Fig. 3(a), the R_0 vs. magnetic field (B) relation exhibits oscillation. Such an oscillation in MWNT has been understood as the AAS effect in a graphite cylinder,[15–17] which originates from phase interference of the electron waves encircling the cylinder in opposite directions, and modulated by magnetic flux enclosed with an oscillation period $\Delta B=(h/2e)/(\pi r^2)$, where r is the radius of cylinder. The inset of Fig. 3(b) shows the ΔB vs r^{-2} relation using the radii measured by HRTEM. It exhibits the linear relation and the order of the slope of 10^{-16}, coinciding with the order of $h/2e\pi$. This is evidence of AAS oscillation and therefore strongly supports that this Coulomb blockade depends on the electron phase interference of the MWNT as the EME.

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[1] G.-L. Ingold and Y.V. Nazarov, in *Single Charge Tunneling* edited by H. Grabert and M.H. Devoret, NATO ASI Series B, Vol. 294 (Plenum, New York, 1991), p. 21.
[2] M.H. Devoret *et al.*, Phys. Rev. Lett. **64**, 1824 (1990).
[3] S.M. Girvin *et al.*, Phys. Rev. Lett. **64**, 3183 (1990).
[4] S.H. Farhangfar *et al.*, Europhys. Lett. **43**, 59 (1998); T. Holst *et al.*, Phys. Rev. Lett. **73**, 3455 (1994).
[5] X.H. Wang and K.A. Chao, Phys. Rev. B **56**, 12 404 (1997); **59**, 13 094 (1999).
[6] P. Joyez *et al.*, Phys. Rev. Lett. **80**, 1956 (1998).
[7] A.O. Caldeira and A.J. Leggett, Phys. Rev. Lett. **46**, 211 (1981).
[8] A.N. Cleland *et al.*, Phys. Rev. Lett. **64**, 1565 (1990).
[9] P. Delsing *et al.*, Phys. Rev. Lett. **63**, 1180 (1989).
[10] L. Langer *et al.*, Phys. Rev. Lett. **76**, 479 (1996).
[11] Y. Imry, *Introduction to Mesoscopic Physics* (Oxford University Press, Oxford, 1997).
[12] E.g., S.J. Tans *et al.*, Nature **386**, 474 (1997).
[13] J. Haruyama *et al.*, Phys. Rev. B **62**, 8420 (2000); D. Davydov *et al.*, *ibid.* **57**, 13 550 (1998); J. Haruyama *et al.*, in *Quantum Mesoscopic Phenomena and Mesoscopic Devices in Microelectronics*, edited by I. Kulik *et al.*, NATO ASI series C, Vol. 559 (Plenum, New York, 2000), p. 145.
[14] A. Bachtold *et al.*, Nature **397**, 673 (1999).
[15] D.Y. Sharvin and Y.V. Sharvin, Sov. Phys. JETP **34**, 272 (1981).
[16] G. Bergman, Phys. Rev. Lett. **48**, 1046 (1982).
[17] Yu.V. Nazarov, Sov. Phys. JETP **68**, 561 (1989).