Intrinsic Nonlinear Conductance of Mesoscopic Conductors

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The conductance of a mesoscopic conductor is shown to be a sensitive function of the dc voltage on the system. In the low-temperature limit, changing the potential drop across the system by an amount on the order of the level spacing of the system is sufficient to change the conductance by e^2/h. Arguments based on scattering theory, analogy with the universal conductance fluctuation, and numerical evidence are presented to support our conclusion. Our theory is in good agreement with the results of harmonic-generation experiments.

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In the mesoscopic regime experiments [1] and theory [2] have convincingly established that the conductance G depends sensitively on the magnetic field, Fermi energy, and impurity configurations [3,4]. It is natural to ask whether G is a similarly sensitive function of the electric field in the system. Such a dependence will manifest itself as a nonlinear conductance of an intrinsic origin (as opposed to one of extrinsic origin such as heating [5] or radiative dephasing [6]). Recent experiments by Webb, Washburn, and Umbach [7] on metal wires and rings and by de Vegvar *et al.* [8] on GaAs wires clearly demonstrate the existence of strong nonlinear conductance. In these experiments a carefully filtered pure harmonic current $I=I_0\cos(\omega t)$ was fed into the sample and the higher harmonics in voltage $V(t)=\sum V_n\cos(n\omega t)$ were monitored. The even harmonics were presumably relatively uncontaminated by heating effects. Three important conclusions emerge from these experiments. (1) The nonlinearity is very strong; even at $I_0=10^{-9}$ A one can still see significant amounts of high harmonics. (2) Depending on the magnitude of I_0, the dependence of V_n on I_0 may belong to one of three different regimes: At the lowest level of the current, $V_n\propto I_0^n$ (henceforth phase I, not seen in the metal experiment), crossing over first to $V\propto I_0$ (phase II) at moderately high current level, and eventually to $V_n\propto I_0^{1/2}$ (phase III, not seen in the GaAs experiment). (3) In the latter two phases $V(t)$ is highly anharmonic, with V_n decaying as a function of n roughly as $1/n$.

These experiments can be understood qualitatively using the theory of Altshuler, Khmelnitskii, and Larkin [9] (AKL) but quantitatively the theory cannot explain the observed strong nonlinearity (see below). In this paper we present a theory which satisfactorily explains the experiments. We further point out that the nonlinearity is a general feature of transport in the quantum regime.

To analyze the experiments, we follow Refs. [7], [8], and [10] and use the model of a current-dependent fluctuating conductance:

$$V(t)=\frac{I(t)}{G_0+(e^2/h)f(I(t)/I_c)}, \qquad I(t)=I_0\cos(\omega t), \tag{1}$$

where G_0 is the conductance of the sample at zero current and e^2/h sets the scale of conductance fluctuation. The function f is dimensionless and describes the variation of conductance as a function of the current. In general f is a highly nonlinear function which fluctuates around zero and whose magnitude is bounded (of order 1). G_0 has been chosen so that $f(0)=0$. We have also introduced I_c to characterize the scale of the onset of strong nonlinearity. For $|x|<1$ (but definitely not for $|x|>1$), $f(x)$ can be adequately described by its linear approximation $xf'(0)$. Apart from these properties f is a sample-dependent function about which few general statements can be made. To analyze the experiments it is best to use a current-dependent conductance; for theoretical discussions a voltage-dependent G is more convenient. While in general a nonlinear inversion is needed to relate one to the other, in the limit where $G_0\gg e^2/h$ we can use $V\approx I/G_0$ for the conversion. This will be done in the following. The reader is invited to look at Fig. 1 where typical examples of the voltage-dependent conductance are displayed. All three phases have been qualitatively reproduced numerically using a current-dependent conductance of the type given in Fig. 1.

In the small-current limit $I_0\ll I_c$, Eq. (1) can be approximated by

$$V(t)=\frac{I_0\cos(\omega t)}{G_0+(e^2/h)f'(0)I_0\cos(\omega t)/I_c}, \tag{2}$$

and it is obvious that the nth-order harmonic $V_n\propto I_0^n$ and that the phase-I behavior is reproduced. On the other hand, if $I_0>I_c$ no simple explicit analytical representation of $V(t)$ is possible. The nonlinearity guarantees the presence of high harmonics, but their magnitude increases only linearly with I_0. This linear dependence stems from the trivial proportionality between V and I. For once $I_0>I_c$, $|f|$ reaches its bound (on the order of 1) and cannot increase further; the nonlinear resistance does not become even more nonlinear with increasing I_0. This is the phase-II behavior. We shall return to the phase-III behavior below.

Thus the crossover point between phase I and phase II gives a good measure of I_c or, equivalently (as long as $G_0\gg e^2/h$, see above), of $V_c=I_c/G_0$. The AKL theory

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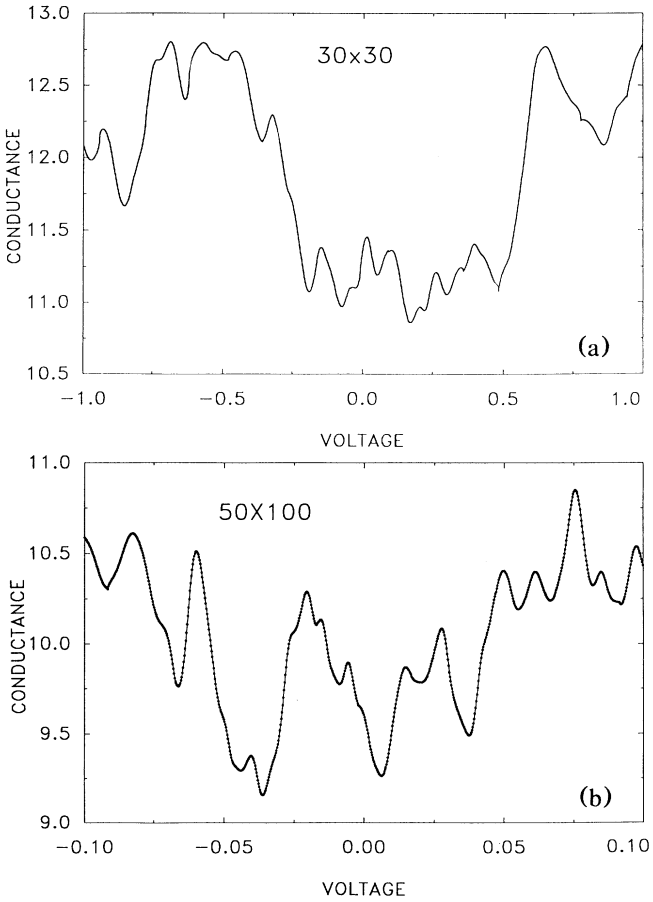

FIG. 1. Dimensionless conductance g as a function of the voltage V. Numerical results from a two-dimensional tight-binding model of size (a) 30 by 30 and (b) 50 by 100 sites. The hopping matrix elements $h_x = h_y = 1.0$, the bandwidth is 8.0, and the average level spacing is (a) 9×10^{-3} and (b) 1.6×10^{-3}.

[9] predicts $eV_c \sim E_T$, the Thouless energy $\hbar D/L^2$, where D is the diffusion constant and L the sample size, or coherence length ξ, whichever is smaller. This prediction is inconsistent with experiment. In the metal experiment [7] (we shall comment on the GaAs experiment [8] below), $1/G_0 = 48.5\ \Omega$; since no phase-I behavior was observed down to $I_0 = 1$ nA, we conclude I_c must be less than 1 nA and therefore $V_c < 5\times10^{-8}$ V. At the same time, E_T was variously estimated to be 2.0×10^{-6} to 1.5×10^{-5} eV. This leads us to believe that the intrinsic nonlinearity is much stronger than predicted by the existing theory.

We propose that the nonlinear conductance becomes observable as soon as δE, the change in electronic energy due to the imposed finite voltage, reaches the level spacing ϵ in magnitude. The crucial difference between our theory and that of AKL [9] is that the latter requires $\delta E \sim E_T = g\epsilon$, where g is the dimensionless conductance $G_0/(e^2/h)$. In a good metal where $g \gg 1$, our theory predicts a much stronger nonlinearity than the AKL model does.

Our proposal is supported by the following argument. The S matrix of a two-lead, N-channel system can be written in the form

$$S = \begin{pmatrix} r_1 & t_{12} \\ t_{21} & r_2 \end{pmatrix}, \tag{3}$$

where r_1, r_2, t_{12}, and t_{21} are all $N\times N$ matrices. There is a powerful argument due to DeWitt [11] relating the S matrix of a system to the exact eigenvalues of the same system confined in a box:

$$-\frac{1}{\pi}\,\mathrm{Im}\ln\det S(E) = -\frac{2}{\pi}\sum_l \theta_l(E) = \frac{\Delta E}{\epsilon}, \tag{4}$$

where θ_l is the phase shift of channel l and ΔE is the shift of energy eigenvalue due to the scattering potential. To avoid confusion, we emphasize that the DeWitt argument involves two related but different systems. The phase shift and the S matrix belong to the scattering problem, which requires an open system with asymptotic regions extending to infinity. The eigenvalues and the level spacing, on the other hand, belong to the same system but with the leads removed and the system completely sealed off. Therefore the eigenvalues are infinitely sharp and without the Thouless diffusion broadening, which measures the diffusion time of an electron through the system. In a real sample the system is open, and the transmission properties are determined by the scattering matrix the properties of which, in turn, can be inferred via DeWitt's argument from the corresponding closed system.

Consider now an infinitesimal change in the scattering potential which may be due to a change in the voltage or a displacement of a scatterer. To first order this shifts the energy levels by δE. Equation (4) indicates that the sum of the phase shifts must change by an amount on the order of $\delta E/\epsilon$:

$$-\pi\delta E/\epsilon = \delta\,\mathrm{Im}\ln\det S = \mathrm{Tr}S^\dagger\delta S$$
$$= \mathrm{Tr}[r_1^\dagger dr_1 + r_2^\dagger dr_2 + t_{12}^\dagger dt_{12} + t_{21}^\dagger dt_{21}]. \tag{5}$$

The dimensionless conductance of the system, $g = \mathrm{Tr}(t_{12}t_{12}^\dagger + t_{21}t_{21}^\dagger)$, will change by

$$\delta g = \mathrm{Tr}[t_{12}dt_{12}^\dagger + t_{12}^\dagger dt_{12} + t_{21}dt_{21}^\dagger + t_{21}^\dagger dt_{21}]. \tag{6}$$

Now, $\delta\,\mathrm{Im}\ln\det S$ and δg are not directly related to each other. This is easily seen, as multiplying the S matrix by a phase factor $\exp(i\alpha)$ changes the determinant but not the transmission; similarly one can change the transmission without changing the determinant. So, at first sight, Eqs. (5) and (6), tantalizingly similar as they are, do not tell us anything. Note, however, that both the determinant and the transmission are functions of all the matrix elements of the S matrix. A $2N$ by $2N$ unitary matrix is parametrized by $4N^2$ real parameters and forms

a $4N^2$-dimensional manifold. To move about this manifold in such a way as to change the phase according to Eq. (5) without changing the conductance according to Eq. (6) requires confining the $4N^2$ parameters to move along a curve. In the absence of any good symmetry this is a highly unlikely event [12]. We therefore conclude that a change in voltage will cause, with probability 1, a change in the conductance. To estimate the magnitude of δg, we note that extrapolating δE to ϵ in Eq. (5) results in a change of the determinant of roughly $\exp(i\pi)$. Since the determinant of a unitary matrix can be written in the form $\exp(i\phi)$, where $\phi \in [0,2\pi]$, this suggests that if $\delta E \sim \epsilon$ the S matrix is substantially altered. Results from the random matrix theory of the universal conductance fluctuations (UCF) [13] lead us to believe that this changes the conductance by e^2/h. Note the correlation between $\delta E \sim \epsilon$ and δg is only a probabilistic one. As a consequence of such a correlation, the conductance depends on the voltage in such a way that every time the latter changes $\delta V \sim \epsilon/e$ the conductance changes roughly by e^2/h.

Our result is new but not surprising. The language we have used provides a unified framework in which to discuss conductance fluctuations. Consider the celebrated example of scatterer motion [3], in which moving a scatterer by a distance $d \sim k_F^{-1}$ leads to a $\Delta g \sim 1$; any change in an external electrical field will disturb the overall scattering-potential landscape and cause the conductance to change. To understand UCF from this point of view, consider a sample of volume Ω in which the electron wave functions are essentially extended. In such a system, moving a scatterer [described by a delta-function potential $U\delta(x)$] by a short distance d is expected to shift the energy level by

$$U(|\psi(0)|^2 - |\psi(d)|^2) \approx U \sin^2(k_F d)/\Omega .$$

Since $U \sim E_F a^3$, where a is on the order of atomic radius, $\Delta E \sim \epsilon \sin^2(k_F d)$. By our S-matrix argument, this causes the conductance to change by $|\Delta g| \sim \sin^2(k_F d)$, as known from the theory of UCF. Likewise, the magnetic fingerprint can be understood using the same language: If the magnetic flux Φ threading a two-dimensional sample of area πR^2 is changed by hc/e, the conductance is expected to change by $\sim e^2/h$. From our point of view, this is because the same change in Φ also changes the single-electron energy by roughly $(e\hbar/mc)\delta\Phi/\pi R^2$, which is on the order of $\epsilon \sim 2\pi\hbar^2/mR^2$. These are all examples of a more general result: Changing the *parameters* of a Hamiltonian such that the energy levels are changed by ϵ generally affects the conductance by e^2/h. On the other hand, the dependence of G on E_F, which is *not* a parameter in the Hamiltonian, is much weaker and is indeed given by $g\epsilon$ and *not* by ϵ. This is due to the fact that $\hbar\,\partial\theta/\partial E$ is the scattering time [14]; equating this to the time needed for an electron to diffuse across the sample reproduces the familiar and experimentally confirmed result $\delta g/\delta E \sim E_T^{-1}$.

A diagrammatic calculation of this nonlinear effect requires considerable care to maintain the small but non-negligible level separation ϵ. Previous theories are based on a quasicontinuous energy spectrum. Within such a theoretical framework only local perturbations (such as the displacement of a scatterer) are picked up; the long-wavelength perturbations typified by an E field will not become visible until the characteristic energy scale reaches E_T. An attempt to evaluate $\langle g(V)g(0)\rangle$ diagrammatically is in progress.

We have carried out extensive numerical calculations to verify our findings. Our technique is identical to that used by Stone [15] in his study of the magnetoresistance fluctuations and is based on calculating recursively the Green's function of the system. The dc electric field was assumed to be uniform and introduced through a linear slant of the on-site potential along the direction of transmission. The transmission at a fixed energy was first calculated. Two typical results are given in Fig. 1. It is clear that (1) the transmission is a complicated function of the voltage similar to the magnetic fingerprint commonly seen in mesoscopic systems; (2) no $V \leftrightarrow -V$ symmetry exists, and the system is rectifying; (3) the conductance fluctuation has a magnitude on the order of 1; (4) the voltage change needed to cause a qualitative change (in the sense of going from a maximum to the next minimum) in transmission is on the order of ϵ, the level spacing (calculated for convenience for a clean system of the same size), and not $g\epsilon$. In Fig. 2 we show the normalized conductance autocorrelation function [16] $C(V) = \langle \delta g(V_0+V)\delta g(V_0)\rangle / \langle \delta g^2 \rangle$. The characteristic voltage where $C(V)$ drops to e^{-1} is marked by A and is seen to be within a factor of 2 of ϵ for the system (marked by C) and much smaller than $\langle g\rangle\epsilon$, marked by B. Note the

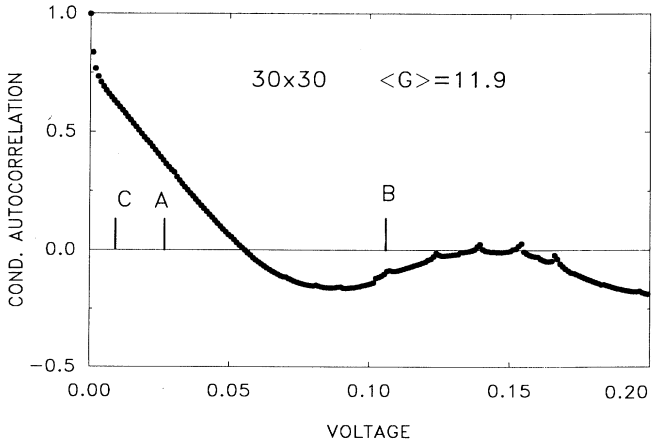

FIG. 2. Normalized conductance autocorrelation function $C(V)$ (see text) for 30 by 30 systems. The voltage at which $C(V)$ drops to e^{-1} is marked by A, and ϵ and $g\epsilon$ by C and B, respectively.

significant negative correlation at large values of V, which we attribute to the existence of important slower fluctuations of g as a function of V. This and a direct inspection of Fig. 1 suggest that the dependence of g on V has rich harmonic contents; while the highest rate of variation is given by ϵ, slower variations, including those on the scale of $g\epsilon$, are also important. We have also carried out simulations with an imposed *transverse* voltage and found a similar effect on the conductance.

So far we have been concentrating on the transmission coefficient at zero energy. If there is a finite voltage difference across the sample, the overall conductance is given by

$$G=\frac{e^2}{h}\int_0^{eV}\frac{dE}{eV}\,\mathrm{Tr}[t_{12}(E,V)t_{12}^{\dagger}(E,V) + t_{21}(E,V)t_{21}^{\dagger}(E,V)]\,. \quad (7)$$

All the transmission amplitudes $t(E,V)$ fluctuate, but the fluctuations of $t(E_1,V)$ and $t(E_2,V)$ become uncorrelated once $|E_1-E_2|$ exceeds E_T (see above); therefore [9] the fluctuation of the overall conductance G gradually diminishes in amplitude like $(E_T/eV)^{1/2}$. We concur with previous authors [7,8,10] and have confirmed using direct Fourier analysis that this leads to the phase-III behavior. In summary, the harmonic-generation experiment is characterized by two different energy scales. The onset of strong nonlinearity takes place at $eV\sim\epsilon$ where phase I meets phase II; the beginning of phase III is at $eV\sim E_T=g\epsilon$. In the GaAs experiment [8], $g<5$, which explains why no serious conflict with earlier theories was found. In the Sb experiment, $g\approx 500$, and phase II and phase III were clearly resolved. Using an estimated value of $E_T=1.2\times10^{-5}$ eV (which is consistent with the estimated level spacing $\epsilon=2.4\times10^{-8}$ eV and a $g=500$), we find the crossover current between phase I and phase II $I_{12}=0.5$ nA and the crossover current between phase II and phase III $I_{23}=250$ nA, in fair agreement with the experiment.

The nonlinearity discussed here should not be confused with the electric-field-induced delocalization. In the weak-localization regime it is known that apart from heating effects the *average* conductance is not affected by a dc electric field [17], a result which is fully consistent with our theory which predicts electric-field-dependent *variations* around the average value.

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