Conductance quantization at room temperature in magnetic and nonmagnetic metallic nanowires

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This paper shows that conductance quantization at room temperature is a physical and reliable observation. This is demonstrated by conductance histograms taking all (12 000) consecutive nanowire conductance experiments in Au at room temperature. On the other hand, conductance curves in Ni, a room-temperature ferromagnet, show staircase-quantized behavior, but the histograms do not show quantized peaks, most probably due to the lifting of the spin degeneracy. [S0163-1829(97)50108-1]

Recent experimental results using scanning tunneling microscopy (STM),[1,2] mechanically controllable break junctions,[3] and quantum table-top experiments using household macroscopic wires[4,5] have shown conductance quantization (CQ) in metallic nanowires at room temperature (RT). Previous to these experiments, CQ was predicted theoretically[6,7] in these nanostructures. Metallic nanowires (NW) are obtained by breaking a contact between two metallic electrodes. The contact does not break cleanly but is stretched into many nanofilaments.[4,5] At the last stages, one thread remains, and quantum-mechanical effects take over. This happens in all the experiments mentioned above.[1–5] If, simultaneously to the formation, stretching, and breakage of the NW, its conductance is measured, a staircase dependence is found just before it breaks. Conductance histograms using up to 100–200 such curves have been reported showing CQ peaks. However, some criteria have been always used to select the conductance curves with which the histogram is built. The metallic nanowire formation has been also predicted by molecular-dynamics simulations,[8] and recent scanning electron microscopy and chemical analysis experiments have displayed it in real time as a macroscopic metallic contact breaks.[5,9]

In this work histograms built using all (up to 12 000), *consecutive* conductance curves are presented for Au, Pt, and Ni nanowires at RT. This is ~100 times more than any previously reported experiments and without histogram sample selection. The conductance histogram for Au at RT and in air shows clear CQ peaks. The histogram for Ni electrodes does not show CQ peaks, even though the conductance shows a stepped behavior, most probably due to the lifting of the spin degeneracy.

The experiments are performed in a home-built STM at RT in air. Two high-purity polycrystalline macroscopic electrodes with a few tens of mV potential difference between them, are brought in and out of contact. The current flowing through is measured with a current-voltage (I-V) converter working at 10^5 gain (100-mV/μA, 3-μs settling time). The position of one electrode is fixed, while the other is moved with a piezoelectric actuator driven with a triangular wave. The electrode speed is controlled by varying the amplitude and/or the frequency of this signal. The output of the I-V converter (the current signal) is measured with a LeCroy 9354AM oscilloscope, with a 500-MHz bandwidth and a 5-G sample/s sampling rate. The current data acquisition is triggered when the current signal crosses a predetermined value with a predetermined slope; in the case of a breaking contact we are interested in signals with negative slope, i.e., the current decreases as the contact breaks. At least 25 k samples per conductance curve are acquired and used to build the histogram. This histogram is *constructed and displayed in real time*. The histogram parameters, center width and number of bins, are adjusted to cover the experimental current window. The raw histogram is lightly filtered, in real time too, in order to avoid fictitious peaks due either to the difference between the vertical resolution of the current window (8 bits=256 levels) and the number of bins of the histogram, or to the digital electronics.

Typical conductance experiments for Au (a) and Ni (b) nanowires are shown in Fig. 1, where the stepped behavior of the conductance is quite apparent. The curves shown look very similar. However, conductance jumps of about e^2/h are observed more often in nickel. These statements are based on the experience of seeing millions of curves for both metals. When reproducible conductance histograms corresponding to these elements are built, Au presents clear peaks [Fig. 2(a)], but Ni does not [Fig. 2(b)]. In all the elements studied with this technique in our group up to date (more than 10) we find steady and reproducible conductance histograms with a few thousand curves. Evidence of this statement is illustrated in Fig. 2(a) where it can be observed that the features of the Au histogram are not modified when the number of samples changes from 3000 to 12 000. In addition, the histograms obtained with our technique, using a few thousand conductance curves, are identical in different locations of the sample. The experimental conditions for both histograms presented in Fig. 2 are identical; the potential difference between electrodes is $V=90.4$ mV and the electrode separation speed is $v=89\,000$ Å/s.

The gold histogram presented in Fig. 2(a) has been obtained by subtracting 490 Ω from the original data. This is due to backscattering of electrons.[10] The value of the resistance quantifies the average disorder in the nanowire under the experimental conditions. Evidence for this statement is twofold: Tight-binding calculations demonstrate that as disorder grows the amplitude of the quantum conductance step decreases.[10] This is displayed in Fig. 3, where histograms corresponding to conductance curves [Fig. 2(a) in Ref. 10] for nanowires with different degrees of disorder are shown. The parameter D in the figure is a disorder parameter. Notice

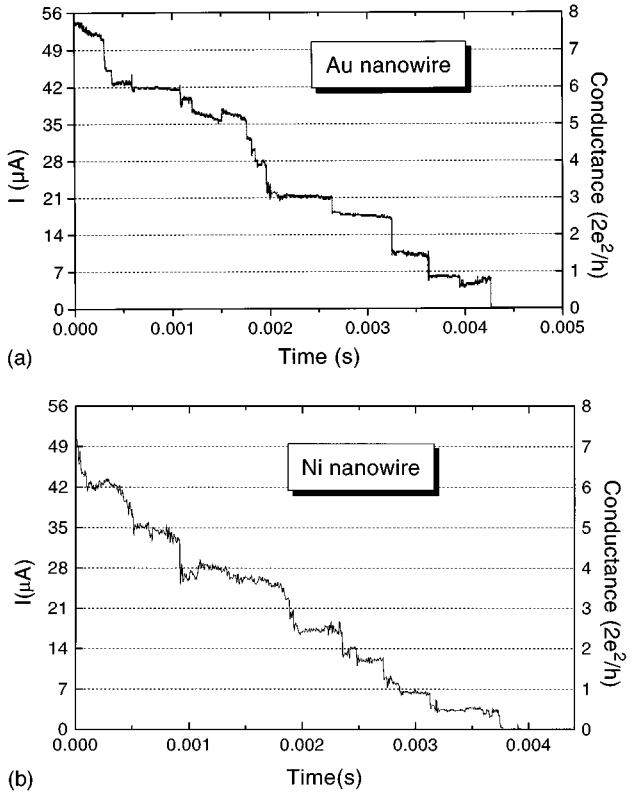

FIG. 1. Typical conductance curves for (a) gold and (b) nickel nanowires at RT in air. The applied potential difference between the separating electrodes is 90.4 mV.

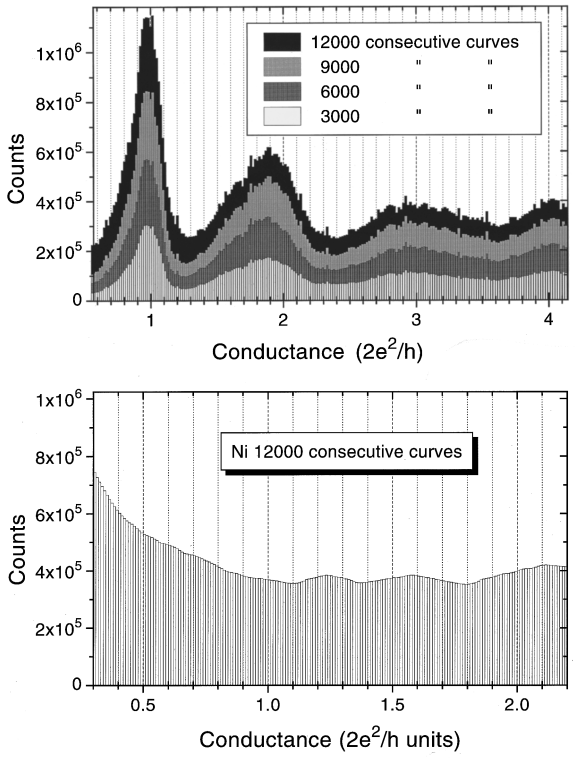

FIG. 2. Conductance histograms for (a) gold and (b) nickel nanowires at RT and in air. The applied potential difference between the electrodes is 90.4 mV. The electrodes separate at 89 000 Å/s. (a) Conductance histogram for gold nanowires. The effect of the number of consecutive samples, $N=3000$, 6000, 9000, and 12 000, is shown. These histograms are obtained after subtracting 490 Ω from the original (see text). (b) Conductance histogram for nickel nanowires built with 12 000 consecutive curves. The applied potential difference between the electrodes is 90.4 mV and the electrodes separate at 89 000 Å/s. No resistance has been subtracted from the original data.

that for zero disorder the conductance follows a perfect $2e^2/h$ step staircase, while for increasing disorder the amplitude of the conductance step decreases. In conductance terms, this means that the position of the peak for the nth channel departs from the value $nG_0=n2e^2/h$ following approximately a law $G_n=nG_0(1-\beta n)$ with β small. Therefore, the effect of disorder, for low-n values, can be quantified by a constant resistance value. This is basically the behavior experimentally found in gold nanowires as shown by the (490 Ω subtracted) histogram in Fig. 2(a). Additional evidence for a conductance that depends mostly on the disorder in the nanowire comes from the electrode separation speed effect on the histogram. This is shown in Fig. 4, where the position of the first peak of the conductance histogram, without subtracting any resistance, is displayed as a function of the electrode separation speed. Notice that as the electrodes separate more slowly, that is, the nanowire stretches more slowly, the position of the first conductance peak approaches the value $G_0=2e^2/h$, corresponding to a perfectly ordered nanowire. The peak position is determined from a Gaussian fit of the data between $0.8G_0$ and $1.2G_0$. The line in Fig. 4 is drawn as a guide to the eye. The experimental findings reported above can be theoretically justified in detail as follows: When considering experiments revealing conductance quantization at RT it is necessary that the mean free path of the charge carriers, l, is larger than the length L and the width W of the constriction, filament, or nanowire that carries the current. Then, using a Landauer formalism,[11] the conductance for a given NW, $G(n)$ can be expressed as

$$G(n)=\frac{e^2}{h}\left(\sum_{i=1}^{N_1} T_{i\uparrow}(n)+\sum_{i=1}^{N_2} T_{i\downarrow}(n)\right), \tag{1}$$

where the sums run over occupied states, $T_{i\uparrow}(n)$ is the transmittance for the ith channel with spin up, and $T_{i\downarrow}(n)$ is the transmittance for the ith channel for spin down for a given nanowire with a certain atomic configuration n. In a metal, the Fermi wavelength λ_F is ~0.5 nm, and the level separation is ~1 eV, which means that at RT conductance quantization can be observed if W and L are smaller than l~10 nm. For diamagnetic nanowires $T_{i\uparrow}(n)=T_{i\downarrow}(n)$. Then Eq. (1) becomes

$$G(n)=\frac{2e^2}{h}\sum_{i=1}^{N} T_i(n). \tag{2}$$

This function has a staircase behavior but the height of the quantum conductance step is $2T_i(n)e^2/h$, not $2e^2/h$. To have a quantum step equal to this last value we should have $T_i(n)=1$. This happens when there is spin degeneration and the conductance is ballistic, i.e., there is no backscattering due to disorder and surface roughness in the NW. In a diamagnetic metal such as Au, there is spin degeneration in the nanowires formed at RT, as demonstrated by Fig. 2(a). For a

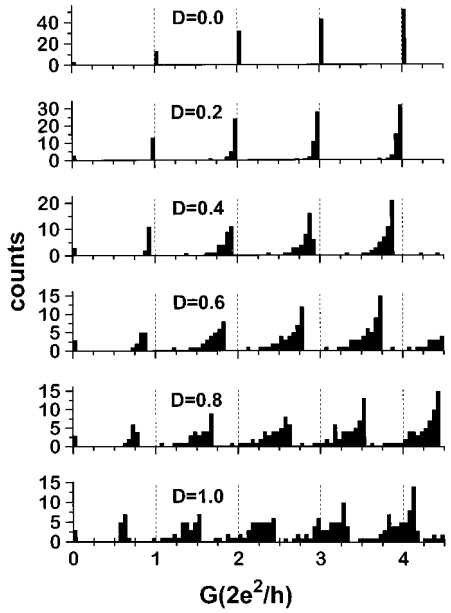

FIG. 3. Histograms corresponding to conductance curves [Fig. 2(a) in Ref. 10] obtained by tight-binding calculations, showing the conductance of a 30×30 nanowire with different degrees of disorder. The parameter D quantifies the degree of disorder.

ferromagnetic metal such as Ni the situation on breaking a contact is considerably more complex. There is a spin-dependent density of states at both sides of the nanowire, and the transmittance should depend, besides topological factors, on the spin direction [Eq. (1)]. In addition, the number of occupied states with spin up (N_1) does not have to be equal to the number of occupied states with spin down (N_2) for any nanowire. Magnetic effects should then play an important role in the NW transport characteristics. In principle, $0<T_{i\uparrow,\downarrow}(n)<1$; in fact, the experiment shown in Fig. 2(b) suggests that $T_{i\uparrow,\downarrow}(n)$ is a random number between 0 and 1 due to the lack of spin degeneration and a transmittance that depends very much on the particular topology of the ith state in the n-NW. This implies that Au histograms show well-defined peaks because $T_i(n)\approx T(n)\approx T$, but for Ni the histogram is flat because there is no spin degeneration of the electronic states in the nanowire and their transmittance depend strongly on the actual topology of the nanowire, as shown in Fig. 2(b). However, the individual conductance curves present well-defined steps.

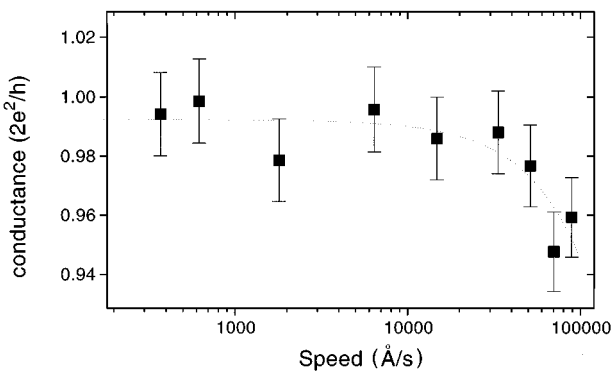

FIG. 4. Dependence of the position of the first conductance peak for gold nanowires on the electrode separation speed. The line is drawn as a guide to the eye.

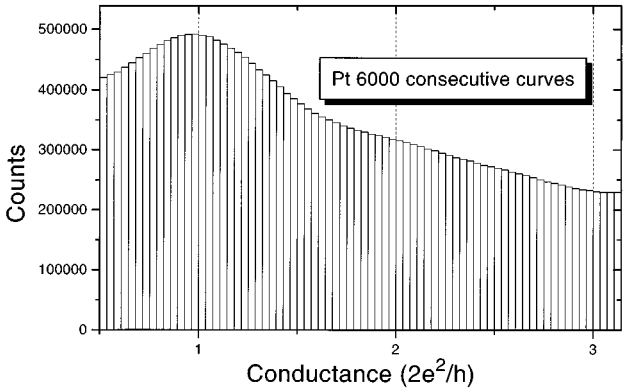

FIG. 5. Conductance histogram for platinum nanowires built with 6000 consecutive curves. The applied potential difference between the electrodes is 90.4 mV and the electrodes separate at 89 000 Å/s. No resistance has been subtracted from the original data.

It could be argued that the flat histogram measured for Ni is just a consequence of a flat distribution of transmittance values due to a different plastic behavior at the nanoscale when compared with Au. In other words, while in Au there is a peaked distribution of $T(n)$'s for the different n nanowires under the experimental conditions obtained, this distribution is broader for Ni, rendering the conductance histogram flat. We have performed additional conductance experiments with Ag, Pt, Co, Fe, and Cu electrodes, and, while Ag, Pt, and Cu show peaks (better defined for Ag and Cu, softer materials) in their conductance histograms, Co and Fe, RT ferromagnets show a flat histogram. There is certainly some effect due to the nanomechanical behavior of the different metals when brought in and out of contact; however, Pt and Ni are very similar metals from the mechanical point of view, but Ni shows a flat conductance histogram and Pt does not (Fig. 5). Relevant data[12] for the polycrystalline materials used in the conductance experiments is shown in Table I. As shown in Fig. 5, Pt shows a broad peak at $2e^2/h$ and another, almost unnoticeable, at $4e^2/h$. Due to the large half-width of the observed peak, it is not difficult to see that a material with the same mechanical properties as Pt, but with a conductance quantum of half the value for spin degeneracy, will produce a flat histogram.

Finally we would like to mention that it is well known that purely geometrical effects (the shape of the nanowire) can produce a widening of the conductance histogram peaks.[13] This effect cannot account for the large peak dis-

TABLE I. Melting temperature and mechanical properties of the metal used in the conductance experiments.

	Melting temperature (°C)	Bulk modulus (GPa)	Tensile modulus (GPa)	Poisson's ratio
Pt	1772	276	170	0.39
Ni	1453	177.3	199.5	0.312
Au	1064	171	78.5	0.42

placement observed experimentally. A combination of this effect with the lifting of the spin degeneracy and the shift of the conductance due to disorder in the nanowire is, in our opinion, what produces a flat conductance histogram for Ni.

Summarizing, conductance histograms taking all (12 000) consecutive nanowire conductance experiments in Au at room temperature show the quantized nature of the transport through nanowires at room temperature. The measured histograms are totally reproducible. Departures from a perfect $2e^2/h$ quantization can be explained in terms of the effect of disorder. On the other hand, conductance curves in Ni, a RT ferromagnet, show staircase quantized behavior, but the histograms do not show quantized peaks most probably due to the lifting of the spin degeneracy combined with the effect of disorder, and, a widening of the peaks due to purely geometrical effects.

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