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LETTER TO THE EDITOR

In situ monitoring of quantum conductance in electrodeposited magnetic point contacts

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Abstract

Electrodeposition of Ni over electrodes initially separated by less than 100 nm has made possible the study of the conductance across increasingly smaller gaps between two magnetic materials. *In situ* measurements of the inter-electrode impedance showed conductance steps at multiples of e^2/h, confirming the absence of spin degeneracy in ferromagnetic nanocontacts. No magnetoresistance larger than 10% for conductance values smaller than $50\,e^2/h$ was observed during the contact growth or dissolution under a sweeping external magnetic field.

The quantum conductance (QC) phenomenon establishes the size limits where the wave nature of electrons is dominant in electric transport. For normal metals, with a typical Fermi wavelength smaller than 1 nm, it has been clearly shown that a contact made of a few atoms exhibits a conductance quantized in units of $2e^2/h$ ($\approx(13\,\mathrm{k}\Omega)^{-1}$), where the factor 2 is related to the spin degeneracy [1]. Ferromagnetic systems are of particular interest in exploring QC as the exchange splitting energy lifting the spin degeneracy should result in e^2/h conductance step values. The first experiments showed indeed that steps at $e^2/2h$ and $3e^2/2h$ are possible [2–4]. A single conductance trace reveals quantum steps, but the sum of several thousands of traces generally show no clear tendency of preferred conduction values [5].

Nanosized magnetic contacts are also attractive for investigating magnetoelectronic effects. The magnetic configuration at the contact is a key parameter for controlling its conductance. Calculations from Imamura *et al* [6] predict that conductance steps at multiples of $e^2/2h$ appear in a ferromagnetic alignment at the contact, and only even multiples should be present if an antiparallel configuration exists. The geometry imposed by a constriction makes an antiparallel configuration within a length scale of a few atoms plausible [7], even though domain walls extend over a much larger width in bulk ferromagnetic materials. The initial investigations of magnetoresistance effects have been very encouraging, given the spectacular results obtained on electroplated contacts in a T-shaped geometry, where the point-contact resistance values change by a factor of 30–40 when a few ohms connection is made between micron-sized

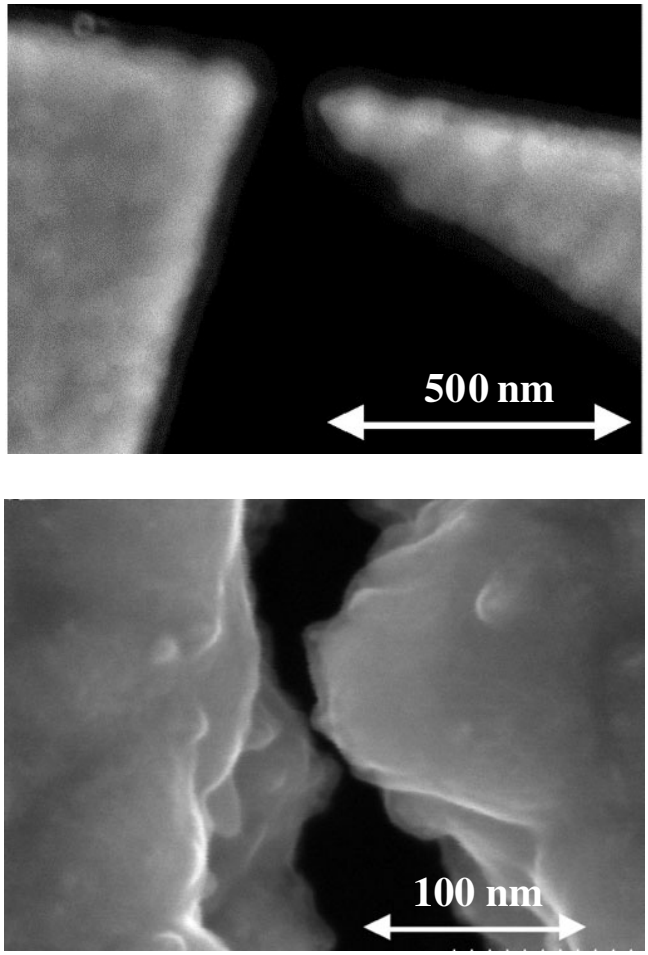

Figure 1. Scanning electron microscopy micrograph showing the separation between patterned Au electrodes after FIB milling (top) and after subsequent Ni electroplating (bottom).

wires [8, 9]. The interface between the two metals is key for electronic transport properties: it is essential to devise experiments keeping control of the surface (electro)chemical state.

We present *in situ* conductance measurements performed on Ni point contacts grown over pre-patterned sub-micron electrodes, separated by a gap smaller than 100 nm. The experiments of Morpurgo *et al* [10], which showed successful QC for Au contacts, are extended to the study of magnetic materials. The main advantage of this technique is the possibility to monitor the conductance of the contact *in situ* during its growth and observe QC over a timescale long enough to sweep an external magnetic field and investigate magnetoresistance properties. The electrochemical growth is a reversible process and dissolution results in the opening of the contact, for which QC can also be observed. In this letter we describe in more detail the electrochemical conditions, as well as the limits of the interpretations of the results. In particular, the importance of patterning on a small scale of the initial electrodes is pointed out. Clear QC can be established for electrodeposited Ni contacts if an external magnetic field is applied to the sample during its growth, but no significant magnetoresistance is found for the presented patterned geometry.

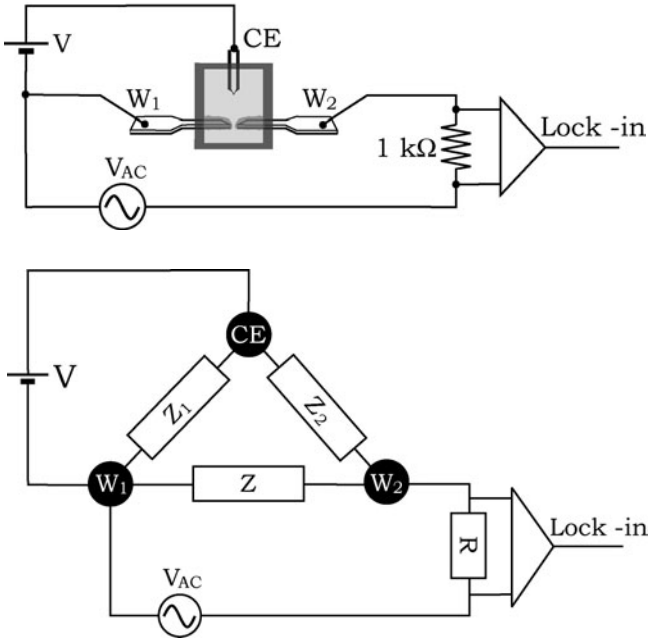

Figure 2. Electrochemical set-up (top) and the corresponding electric circuit (bottom).

Initial patterning is obtained by photolithography, usually combined with e-beam lithography, or focused ion beam (FIB) milling in our case. This is a versatile procedure allowing us to modify the shape of the connection during the milling process. Lines of 1–2.5 μm width were patterned by photolithography on Si/SiO_2 (300 nm)/Ti (10 nm)/Au (100 nm) wafers. FIB milling was then used to cut the line, leaving gaps of typically 100 nm width, and shaping the two electrodes (W_1, W_2) with a flat or arrow end (figure 1). Over-cuts of the SiO_2 ensured the absence of leakage current due to metal ions implanted during the FIB milling process. No current exceeding 10 pA was detected within 2 V voltage difference between W_1 and W_2. Electrodeposition of the metal of interest over the two electrodes, following the scheme of figure 2, permitted us to slowly close the gap. A potentiostat ensured an adequate current to flow between CE and W_1, keeping the potential difference between W_1 and the reference electrode controlled (the reference electrode is not indicated in figure 2). The impedance Z between W_1 and W_2, which is the quantity of interest, was monitored through the series resistor R and the small AC excitation V_{AC} by lock-in detection. There are several key precautions taken to avoid experimental bias in measurements. To ensure that the monitoring current passes mostly through the impedance Z under investigation, a large value for the bath impedance Z_2 was needed (the potentiostatic control inhibits the AC current path through Z_1). Low frequencies for the monitoring AC voltage were necessary to avoid shorting of the AC current by the capacitive part of the impedance of the electrolytic bath. Impedance spectroscopy prior to plating showed typically 1 MΩ values at frequencies lower than 10 Hz, becoming smaller than 100 kΩ at 100 Hz. Another more severe limitation was caused by the intrinsic ionic conductivity of the bath. At a typical conductivity value of 1 Ω^{-1} m^{-1}, two electrodes with disc-shaped ends of 100 nm diameters separated by 1 nm can be shorted by a resistance of the order of 10^5 Ω. Therefore, good sub-100 nm patterning techniques were necessary for the initial electrodes. Furthermore, interpretation of the resistance between

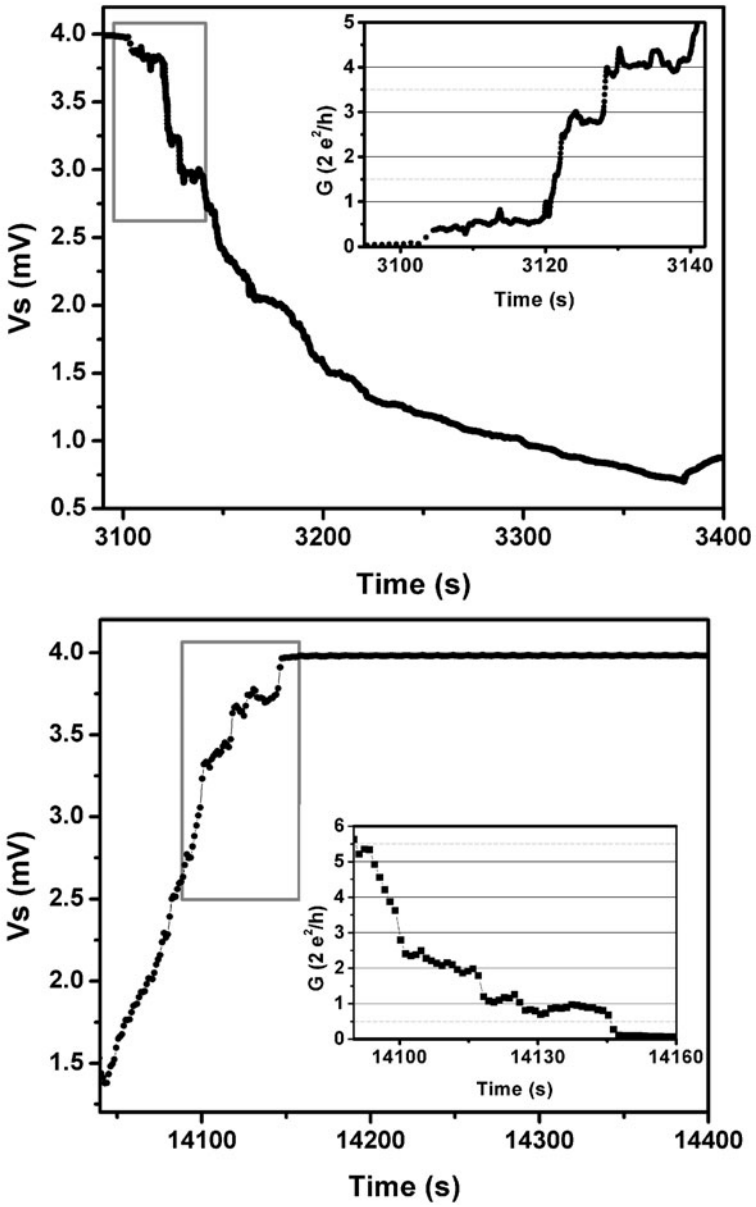

Figure 3. Rms voltage between the two electrodes as a function of time. Insets indicate the corresponding conductance showing steps at multiples of e^2/h during the opening of a contact (top) and $2e^2/h$ during the contact closure (bottom).

non-connected electrodes in terms of tunnelling is inadequate, and an error of several per cent in measuring the first conductance steps is possible. Finally, the amplitude of the monitoring AC excitation must be small enough. For slow deposition, the applied potential is at the onset of the reduction potential of the metallic ions in solution. Following the Nernst law [11], the current varies exponentially around this value, which implies that changing the voltage

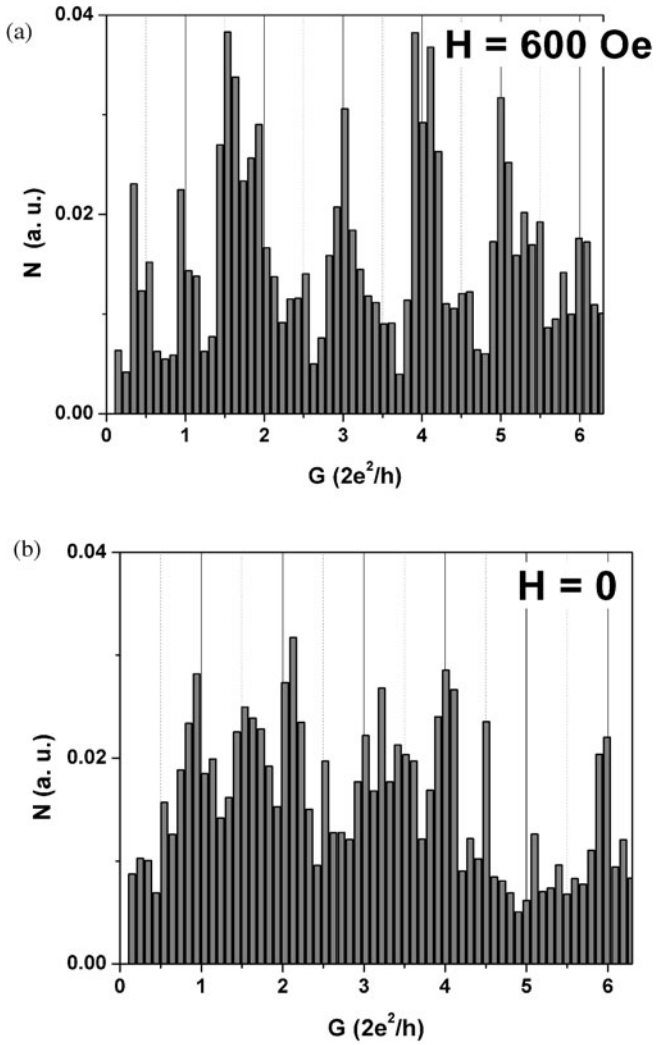

Figure 4. Histograms of the time traces of the conductance of 20 samples, showing the difference in statistics between contacts made under applied field (top) or without applied field (bottom).

by several millivolts modifies drastically the electroplating rate. Our experience is that the monitoring AC rms amplitude should not exceed a few millivolts.

We reproduced the results of Morpurgo *et al* [10] and QC was clearly observed for Au electroplating. Ni electrodeposition was obtained using a Ni sulfamate bath, optimized for minimizing deposit strains and the granularity of the plating. Atomic force microscopy and scanning electron microscopy showed that the typical grain size does not exceed 5 nm. We found, however, that the interface between the two electrodes can show larger irregularities, reaching 20 nm size (figure 1). We also observed a slower deposition rate between the electrodes than over the patterned electrodes. The ion depletion near the electrodes is likely responsible for that effect. The time monitoring results for Ni plating are shown in figure 3. Quantum steps at odd and even multiples of e^2/h are found. The occurrence of quantum steps

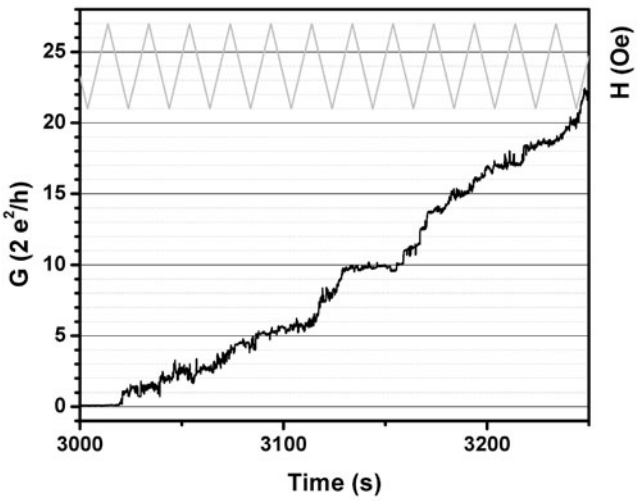

Figure 5. Time trace of a closure of a Ni contact (4 mV rms at 200 Hz), under swept applied field (top grey curve) of 600 Oe amplitude. The resistive part of the trace is indicated, with conductance values under $5e^2/h$ significantly modified (up to 50%) by the capacitive part of the circuit.

was rarely observed for samples grown and dissolved without applying an external magnetic field (reaching 600 Oe in our set-up). Such a tendency is illustrated in the histograms of figure 4. Occurrence of quantum steps is more clearly shown by summing the time traces of the conductance for 20 measurements. Peaks at several multiples of e^2/h appear, with a higher occurrence of even multiples. However, for samples made without applying an external magnetic field, a histogram with the same number of samples and normalization factor revealed a much less pronounced structure. This confirms the idea that a random magnetic orientation at the contact makes the conductance time trace less reproducible [5].

Our attempts to find evidence of changes of QC under applied magnetic field did not reveal any significant magnetoresistance effects. Measurements were performed on samples shaped with an arrow facing a flat electrode, or two facing arrows, under applied field perpendicular to the substrate. This geometry should facilitate a domain wall pinning at the nanocontact region. Higher AC excitation frequency was necessary to ensure enough significant data points for hysteresis curves, at the expense of current leakage through the capacitive part of the circuit. We found that this loss was $<10\%$ of the signal for conductance values larger than $5e^2/h$ at 200 Hz excitation frequency. No evidence of magnetoelastic or torque effects exerted by the external field on the nanojunctions was found, but the sweeping magnetic field affected the stability of contacts with low conductance values (less than $5e^2/h$). Approximately half of the samples showed abrupt irreversible opening of the contact, and subsequent electrodeposition was necessary to retrieve finite conductance values. We did not observe any significant magnetoresistance up to conductance values of $50e^2/h$ (figure 5) for several tens of contacts studied. This is in strong contrast with the reported very large values obtained for larger initial electrodes [8, 9]. Our Ni nanocontacts must have small sizes given the occurrence of QC, and have surface chemistry purity ensured by the potentiostatic control during resistance measurements. Absence of a domain wall at the nanocontact position is unlikely to happen for *all* samples, and even partial antiparallel alignment of the electrodes should result in (reduced) magnetoresistance effects. Our measurements clearly show that a model of spin-dependent

QC cannot account for the large magnetoresistance effects observed on magnetic contacts not exceeding a few tens of ohms resistance. Models involving magnetoelastic properties, surface modifications after electroplating or mechanical effects should be used to explain the spectacular magnetoresistance values observed by others [8, 9].

In conclusion, we successfully observed QC in Ni electrodeposited nanocontacts, where all multiples of e^2/h appear when a parallel magnetic configuration is constrained by an external field. In the absence of a magnetic field, the occurrence of QC is significantly reduced, especially for the odd multiples of e^2/h. These results confirm the findings on mechanical contacts [3, 5] and show that small magnetic contacts can be obtained by the presented electrochemical method. The possibility to make point contacts in a slow and soft manner is of interest for fundamental studies of magneto-electronic properties. For example, a bipotentiostatic control would allow different voltages to be set for the two working electrodes, allowing the study of heterogeneous magnetic systems. The occurrence of QC is an unambiguous indication of a ballistic transport regime, for which no significant magnetoresistances were found in our samples.

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