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Conductance quantization in magnetic nanowires electrodeposited in nanopores

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Magnetic nanocontacts have been prepared by a templating method that involves the electrodeposition of Ni within the pores of track-etched polymer membranes. The nanocontacts are made at the extremity of a single Ni nanowire either inside or outside the pores. The method is simple, flexible, and controllable as the width of the constriction can be varied reversibly by controlling the potential between the electrodeposited nanowire and a ferromagnetic electrode. At room temperature, the electrical conductance shows quantization steps in units of e^2/h, as expected for ferromagnetic metals without spin degeneracy. Our fabrication method enables future investigation of ballistic spin transport phenomena in electrodeposited magnetic nanocontacts. [DOI: 10.1063/1.1503400]

The investigation of electron transport through metallic nanostructures like nanowires or nanoconstrictions is of great interest in both fundamental quantum physics and nanotechnology. It has been recently demonstrated that atomic scale constrictions in metals can be made by controlling the potential between pairs of electrodes immersed in an electrolytic solution.[1–4] By monitoring the electrical resistance between the two electrodes during the electrodeposition process, a precise control of the constriction has been achieved. One interesting feature of the electrochemical method is that the width of the constriction can be varied reversibly by slowly dissolving metal atoms or redepositing them onto the constriction. Moreover, this method presents several advantages compared to more sophisticated methods like scanning tunneling microscopy (STM) and mechanical break junctions[5] in terms of ease, fine control of the constriction width, reversibility of the process, and long-term stability. Quantized conductance in units of $2\,e^2/h$ have been observed in a few room-temperature (RT) electrochemical experiments involving nonmagnetic metals such as Cu and Au.[1,2] Such steps in conductance appear in ballistic transport when the size of the constriction is of the order of the Fermi wavelength, i.e., of atomic dimensions in a metal.

Recently, there have been experimental investigations of quantum transport in atomic-size contacts of ferromagnetic metals made using different methods such as STM,[6,7] contact between macroscopic wires,[8,9] and break junctions.[10] Quantization in conductance has been reported but the unit of quantization in magnetic materials still remains unclear. Indeed, quantized conductance in units of $G_0 = e^2/h$ are expected for ferromagnetic metals without spin degeneracy. Moreover, the plateaus in conductance are expected to depend on the relative orientation of magnetizations on both sides of the nanocontact. From experimental results obtained with break junctions of ferromagnetic Ni wires, Ono *et al.*[10] have observed that the conductance is quantized in units of $2\,e^2/h$ in a zero-magnetic field and that it is switched to e^2/h if a magnetic field is applied to saturate the magnetization. In contrast, recent low-temperature STM experiments on Fe nanocontacts[7] have shown conductance jumps of e^2/h in the absence of a magnetic field. Besides the observation of conductance quantization, spin dependent ballistic transport effects remain to be studied. For instance, the presence of a domain wall at the nanoscale magnetic constriction,[11] its effect on the number of transmitting channels[12] and large RT ballistic magnetoresistance in small Ni contacts[9] are examples of interesting properties of magnetic nanocontacts. Further studies will require a very fine control of the nanocontact size and magnetization.

In this letter, we report on RT conductance quantization in electrodeposited ferromagnetic nanocontacts. The nanocontacts are made by electrodeposition and dissolution of Ni nanowires fabricated in nanopores. The nanoporous media consist in 22 μm thick track-etched polycarbonate membranes, with fixed pore diameters from 60 to 120 nm and pore densities of about $10^8/\mathrm{cm}^2$.[13] The bottom side of the membrane is covered by electron-beam evaporation with a thick gold layer which plays the role of the cathode for nanowire electrodeposition. On the top side, another gold electrode is deposited through a metallic mask. These two conducting layers serve for the resistance measurement of the nanocontact as explained next. The electrodeposition is performed at a constant plating current (1–10 μA) in an aqueous solution of 0.5 M $NiSO_4$ and 0.3 M H_3BO_3, using a Pt counter electrode and a Ag/AgCl reference electrode [Fig. 1(a)]. The galvanostatic electrodeposition method is used for its advantage of better control of the deposition rate. Two different configurations of the nanocontact have been realized. In configuration A [Fig. 1(b)], the top gold strip is thick enough to completely cover the pores. Prior to the electrodeposition of the Ni nanowires in the pores, the top gold layer is electrochemically covered with Ni in order to establish a purely ferromagnetic nanocontact. The nanocontact is then formed when the first nanowire emerges on the surface at the border of the top Ni electrode. In configuration B [Fig. 1(b)], the top Au electrode is evaporated on all of the surface

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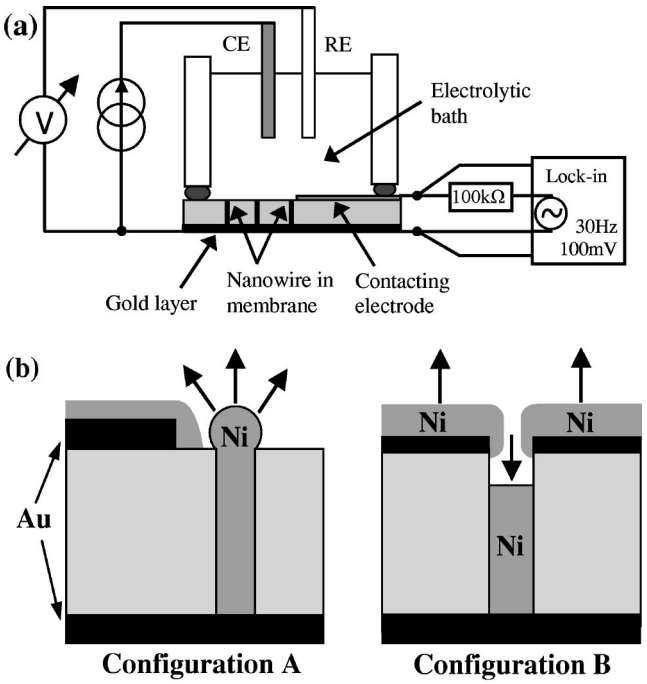

FIG. 1. (a) Schematic drawing of the experimental setup. A Teflon cell is used for the electrodeposition process in the nanoporous membrane. A lock-in amplifier monitors the resistance between the two electrodes on both sides of the membrane. A counter electrode and a reference electrode are used for electrodeposition. (b) Schematics of the formation of nanocontacts using nanoporous membrane. In configuration A, the nanocontact is formed between a growing Ni nanowire and the upper electrodeposited Ni film. In configuration B, the nanocontact takes place inside the pore via Ni deposition on the upper Au electrode.

exposed to the electrolyte solution. However, this layer is thin enough and does not plug the pores so that the electrolyte can penetrate them. The growth of Ni nanowires is interrupted before complete filling of the pores. The nanocontact is finally formed inside the pore through Ni electrodeposition on the top Au electrode. The extremity of the contacted nanowire is very close to the surface prior to the final electrodeposition step. During the electrodeposition process, the two layers are connected to a lock-in amplifier and the resistance between them is measured at 30 Hz with a bias voltage of 100 mV and a resistance of 100 kΩ [Fig. 1(a)].

When the gap between the two electrodes is open, the measured resistance reflects the electrolyte resistance (typically in the range 100 kΩ–1 MΩ). When the width of the gap decreases, the monitored conductance increases drastically, which may result from the electron tunneling across the gap.[3,4,14] Upon making contact, the conductance jumps in discrete steps as the first atoms bridge the gap. The inset of Fig. 2 shows an example of the time variation of the conductance in a zero-magnetic field during the deposition process. The stepwise change in the measured conductance comes from the increase of conducting channels as the width of the constriction increases. At the end of the deposition, the measured resistance is that of a single Ni nanowire, i.e., of the order of 10^2 Ω for the studied diameters. Although steps in conductance appear in the deposition regime, well-defined plateaus, stable up to a few minutes, have been observed during the dissolution. The latter has the advantage that it is possible to slow down the dissolution rate by applying a very

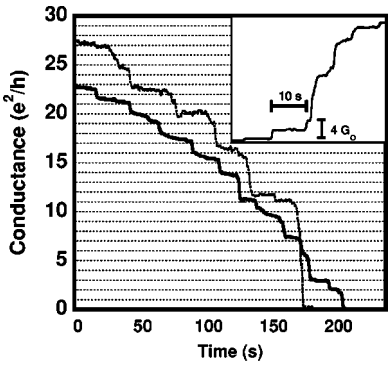

FIG. 2. Conductance steps obtained during the formation of a Ni nanocontact by electrochemical dissolution in configuration A (solid line) and configuration B (dotted line). The inset shows an example of the conductance vs time curve obtained during the electrodeposition process. The resistance of the wire in series with the nanocontact has been subtracted from the total measured resistance.

small current (10 to 100 nA) between the two electrodes. It is noted that as the gold sublayer is stable in the electrolyte solution, the break off concerns only the Ni deposit at the surface of the membrane and likely takes place close to the junction location. As shown in Fig. 2, conductance decreases as a stepwise fashion, from 24 G_0 to values smaller than G_0 in a typical time scale of 200 s. The results show the presence of plateaus at odd values of G_0, indicating that the spin degeneracy has been removed. Similar results have been obtained in configuration B. It should be emphasized that this deposition/dissolution process can be repeated many times on the same sample.

Figure 3 shows the conductance histogram plotted from measurements made on 25 different samples and a total of 120 experiments. All integer values of G_0 present peaks in the histogram, but the peaks at odd values are often less pronounced than those at even values. We also note the presence of peaks at noninteger values of G_0. Several factors can contribute to this behavior, such as the formation of several nanoconstrictions in series during the dissolution process, possible uncertainty in the estimation of the wire resistance, and the ionic conduction of the electrolyte. However, the

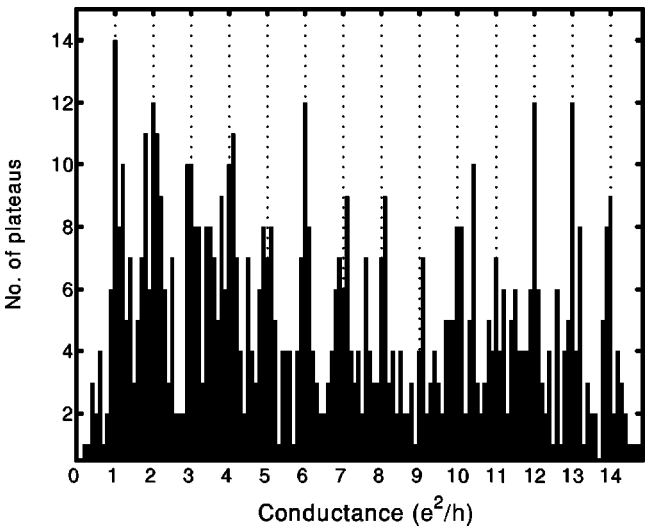

FIG. 3. Conductance histogram of Ni nanoconstrictions obtained from 120 conductance curves presenting at least one plateau, in the absence of a magnetic field. The nanowires diameters range from 60 to 120 nm.

latter is significantly reduced in our templating methods thanks to the small section of the nanowires and the available low pores densities (as small as $10^5/cm^2$). It should be added that measurements have also been made in presence of a magnetic field up to 0.6 T transverse to the wire axis, but no difference has been observed in the saturated state.

In conclusion, we have reported on an original electrochemical method for making magnetic nanocontacts between a Ni nanowire electrodeposited in a porous membrane and a Ni electrode. The method is simple, flexible, inexpensive, and does not require the use of lithography and related techniques. Furthermore, it is possible to extend the method to any metal which can be electrodeposited. The nanocontact is formed at the extremity of a single nanowire of selected diameter growing in a well-defined direction by controlling the electrochemical deposition and dissolution processes. Conductance quantization in units of e^2/h have been observed at RT, using two contact configurations in both remanent and saturated states. Moreover, the nanowire has a well-defined magnetic anisotropy and is protected from dissolution by the polymer. Therefore, such a system might be useful for future investigations of ballistic spin transport through electrodeposited nanoconstrictions and tunneling through controllable magnetic nanogaps.

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