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4-12-2004

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Yang, C-S; Zhang, Chunjuan; Redepenning, Jody G.; and Doudin, Bernard, "*In situ* magnetoresistance of Ni nanocontacts" (2004). *Faculty Publications -- Chemistry Department*. 5.

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In situ magnetoresistance of Ni nanocontacts

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(Received 6 November 2003; accepted 19 February 2004)

Magnetoresistance properties of Ni nanocontacts in the ballistic quantum regime are investigated *in situ* during closure and opening of electrochemically grown planar electrodes. The magnitude of conductance change when sweeping the magnetic field is of the order of the quantum conductance e^2/h for conductance values spanning 1–100 quanta. The relative orientation of electric current and applied magnetic field changes the magnetoresistance sign, with symmetry properties reminiscent of bulk anisotropy magnetoresistance. *Ex situ* investigations of samples of higher conductance values, of the order of 1000 quanta, unambiguously show the analogy with bulk anisotropy magnetoresistance. © 2004 American Institute of Physics. [DOI: 10.1063/1.1705723]

There is a growing interest in making smaller materials for magnetoelectronic applications.¹ The sub-10 nm size opens the possibility to reach dimensions of magnetic electric contacts smaller than the electron mean free path, making possible the study of ballistic transport. Recent reports of spectacular magnetoresistance (MR) in electrodeposited Ni contacts attracted significant interest in the scientific community.^{2–4} However, initial results on mechanical break junction samples indicated much lower MR values.⁵ A consensus for explaining the experimental discrepancies is lacking, and there is a strong need for investigating samples bonded to a surface, minimizing magnetomechanical properties.

If the lateral dimension of an electric contact is smaller than the Fermi wavelength, Landauer formalism predicts a conductance written in the form⁶

$$G = \frac{2e^2}{h} \left(\sum_i^N T_i \right). \quad (1)$$

There is now a full body of experimental work showing that the observed plateau of conductance can be related to Eq. (1) with a preferred occurrence for integer multiples of $2e^2/h$.^{7,8} The estimated Fermi wavelength for ferromagnetic metals is of a few angstroms, and junctions exhibiting such quantum conductance (QC) effects must therefore be of sub-nanometer dimensions.⁹ Investigations of the MR properties of junctions of this size face two difficulties. First, the synthesis should allow stabilization of the QC regime for a timerange long enough for sweeping an external magnetic field and recording the resistance curve. Second, as displacements of a fraction of nanometer are expected to significantly modify the electric conduction properties of the contact, the samples should be designed in a robust way. Minimizing

mechanical strains caused by magnetoelastic properties of the magnetic leads, torque on the sample due to the external field, or magnetostatic interactions when changing the magnetization configuration around the nanocontact are key for obtaining convincing and reproducible experiments.

We present results on MR properties in junctions in the ballistic quantum regime of conduction on planar samples, combining patterning and electrochemistry synthesis methods. Contacts are made by electrochemical deposition and dissolution techniques, with *in situ* investigation of the MR properties. A planar geometry is more suitable for future integration as devices, and allows optimizing the mechanical stability by rigidly binding the sample to the substrate. Electrochemical control ensures that the surfaces are free of oxides.

Initial planar electrodes of 2–5 μm width, and separated by less than 100 nm, were patterned from a Si/SiO₂/Ti(10 nm)/Au(100 nm) wafer using optical lithography and focused ion beam (FIB) milling (Fig. 1). Initial electrodes were patterned in the shape of two facing arrows, or one arrow facing a perpendicular electrode (T shape). Applied dc potentials between counterelectrode (Ni wire) and working (patterned Au) electrodes allow control of the deposition or dissolution rate of Ni. A length of approximately 200 μm of the Au wires was covered, resulting in Ni planar electrodes of shapes similar to the patterned Au substrate, and of approximately 100 nm thickness. The Ni sulfamate electroplating bath $\{[\text{Ni}(\text{SO}_3\text{NH}_2)_2] \text{ 1.5 M, } (\text{H}_3\text{BO}_3) \text{ 0.6 M.}\}$ was optimized for minimizing strain and grain size of the films. The impedance between the two patterned electrodes was monitored through a series resistor R (1 k Ω) under ac excitation of less than 4 mV_{rms} amplitude between the two patterned electrodes. A similar setup was successfully used to make Au nanocontacts¹⁰ and gaps between Pt electrodes.¹¹ Technical precautions necessary to ensure a limited shunting of the monitoring impedance through the electrolytic bath were de-

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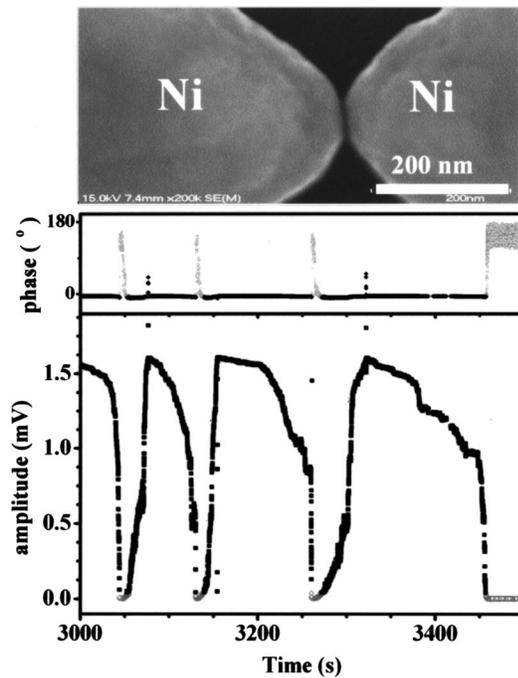


FIG. 1. Top: Scanning electron micrograph of the Ni electrodes after deposition, with a 5 nm gap left intentionally for illustration purposes. The grain size of the deposit is less than 10 nm. Bottom: Time trace of lock-in detection showing raw amplitude and phase data. The clear dots correspond to impedance larger than 100 k Ω .

tailed in a previous article.¹² The phase of the lock-in detection unambiguously confirms the resistive nature of the contacts, in strong contrast with the noisy capacitive component found when the impedance of the system is larger than typically 100 k Ω , values at which the electrochemical bath contributes significantly to the measured impedance (Fig. 1). Such confidence in the resistive nature of the sample is obtained if the lock-in frequency is kept under 200 Hz, therefore limiting the time interval between two measurements (typically 10 ms). The occurrence of QC is confirmed by the statistical analysis over several conductance traces, giving indications that conductance values in multiples of e^2/h are statistically favored, even though individual traces do not exhibit clear quantized values. The spin degeneracy, reflected by the factor of 2 in Eq. (1), is lifted in Ni,¹² as expected from the occurrence of exchange energy, and in agreement with previous experiments.^{13,14}

A plateau of conductance lasting several seconds, and obtained under adequate tuning of deposition or dissolution potentials, allowed sweeping of an external magnetic field and recording of a MR curve (Fig. 2). The field was applied in the plane of the pattern, either longitudinal or transverse to the arrow-shaped electrode(s), or perpendicular to the wafer plane. The magnetic field was swept at rates between 0.1 and 0.5 Hz, with a maximum field amplitude of 0.6 T. Two consecutive different conductance measurement points were typically separated by a magnetic field increment of 2 mT. Time traces of conductance under sweeping magnetic field (Fig. 2) clearly show that the field modifies the conductance in the quantum ballistic regime. For conductance values between e^2/h and $100e^2/h$, the MR is related to a change ΔG , which varies slowly with conductance values. This amplitude is found of the order of e^2/h in most cases (30 samples, with

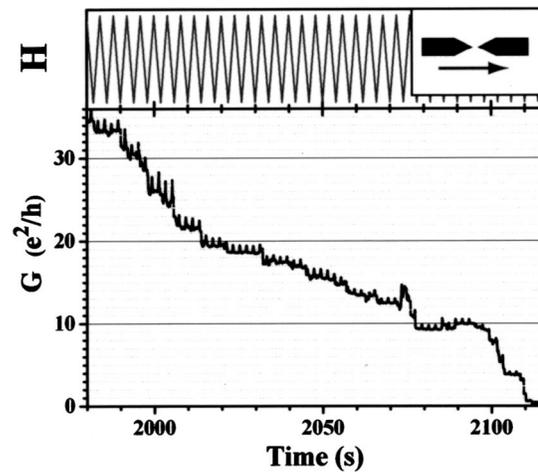


FIG. 2. Time trace of the conductance during opening of a Ni contact, under sweeping applied magnetic field. The field is parallel to the Ni/Au electrodes direction (see inset).

at least five contact closure and openings per sample), and sometimes as small as $0.2e^2/h$. Even though ΔG values are not perfectly reproducible from sample to sample, the trend of ΔG slowly diminishing with G is systematically found.

The sign and type of MR observed change drastically when modifying the relative orientation of the magnetic field and electric current (Fig. 3). However, no noticeable differences are observed when modifying the shape of the initial electrode pattern. The sign of MR for perpendicular orientation is opposite to the one in the longitudinal orientations. This latter case systematically shows much less noisy features, indicating that the fluctuations in the MR data are essentially intrinsic to the sample. More remarkable is that no significant MR is observed when the applied field is trans-

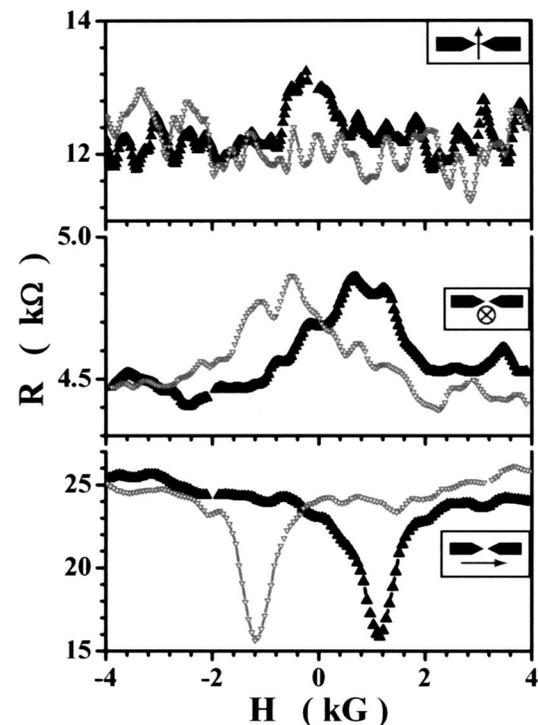


FIG. 3. MR curves of Ni nanocontacts for (top) transverse, (middle) perpendicular, and (bottom) longitudinal orientation of the magnetic field with respect to the Ni/Au electrodes, as indicated by inset diagrams.

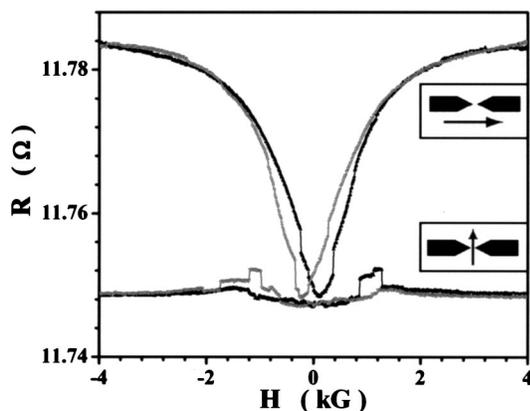


FIG. 4. Magnetoresistance of a low-conductance Ni nanocontact, measured *ex situ*, with applied field transverse and longitudinal to the Ni/Au electrodes.

verse to the contact, even for T-shaped connections, with applied field parallel to the unpatterned electrode (inset of Fig. 3). Spikes in the time trace of the conductance can be found, but at irreproducible applied field values and of irreproducible amplitude. The maximum observed MR is of the order of 70% for conductance values near the single quantum conductance e^2/h (Fig. 3).

Samples of conductance of the order of 10^3 quanta can be taken out of the electrolytic bath for *ex situ* measurements without noticeable change of resistance values. Their resistance value (10–50 Ω) is significantly higher than the electrodes resistance (less than 0.5 Ω). High-fields measurements (up to 7 T) confirm the *in-situ* measurements, as well as the near saturation of the MR at 0.5 T. Samples of a few tens of ohms show around 0.5% MR, corresponding to ΔG of several e^2/h , confirming the trend observed during *in situ* measurements of samples of less than $100e^2/h$ conductance. The MR curves (Fig. 4) are very similar to those of nanocontacts of much lower conductance values, and are strongly reminiscent of typical anisotropy magnetoresistance (AMR) curves, especially when contemplating the difference in high-field conductance values for the two orientations of the magnetic field. The data of Fig. 4 confirms the smaller MR observed in the transverse case, where the magnetic moments in the nanocontacts region deviate little from the transverse orientation when sweeping the magnetic field. Measurements of the perpendicular orientation performed on other samples show that the change of the MR sign can be attributed to a difference in high-field resistance values, confirming the occurrence of AMR effects in our samples.

In conclusion, we successfully made reproducible *in situ*

observations of both of QC and MR in electrodeposited Ni nanocontacts. The change of conductance when stabilizing a nonparallel magnetization state at the nanocontacts is nearly invariant for several orders of magnitude of the conductance values, and the maximum MR is therefore observed around the single quantum of conductance. We find that the conductance change of the order of e^2/h occurs when the magnetization orientation changes from longitudinal to transverse orientation with respect to the current direction. Our findings are similar to those of Viret *et al.*⁵ on mechanical break junctions. For samples of much higher conductance reaching the diffusive regime of conduction, similar changes of conduction with applied magnetic field are found with a behavior similar to bulk AMR. The samples were designed for minimizing the influence of magnetostatic and magnetostrictive effects. One should note however that they do not fully allow to rule out their influence, in particular, if the ductile properties of Au allow the motion of small electrodes.¹⁵

This research was supported by the MRSEC Program of the NSF (DMR-0213808), the Office of Naval Research (N00140210610), the Keck Foundation, and the Nebraska Research Initiative. We thank Mark Johnson (ONRL) for fruitful discussions and help for starting up this project, as well as Professor S.-H. Liou (FIB) and Professor N. Ianno (photolithography) for their laboratory hospitality.

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