Evidence for Spontaneous Spin-Polarized Transport in Magnetic Nanowires

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The exploitation of the spin in charge-based systems is opening revolutionary opportunities for device architecture. Surprisingly, room temperature electrical transport through magnetic nanowires is still an unresolved issue. Here, we show that ferromagnetic (Co) suspended atom chains spontaneously display an electron transport of half a conductance quantum, as expected for a fully polarized conduction channel. Similar behavior has been observed for Pd (a quasimagnetic 4*d* metal) and Pt (a nonmagnetic 5*d* metal). These results suggest that the nanowire low dimensionality reinforces or induces magnetic behavior, lifting off spin degeneracy even at room temperature and zero external magnetic field.

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It is expected that the new generation of devices will exploit spin dependent effects, what has been called "spintronics" [1]. In this sense, the role of low dimensionality in the magnetic properties of materials will become a fundamental issue for combining the standard miniaturization of microelectronics and spin phenomena. From a practical point of view, spintronic devices must exploit different quantum properties without the need of cryogenic temperatures. This fact immediately renders metal nanowires (NW's) a very attractive system because they show quantum conductance effects at room temperature [2]. From an experimental point of view, NW's can be easily generated by putting in contact two metal surfaces, which are subsequently pulled apart; during the elongation, the conductance (G) displays flat plateaus and abrupt jumps of approximately a conductance quantum $G_0 = 2e^2/h$, where e is the electron charge and h is Planck's constant [2]; the factor 2 is due to the spin degeneracy. Recently, this kind of study has led to the discovery that the ultimate NW's show a structure of a suspended linear chain of atoms (LCA) [3–5], whose conductance is equal to $1G_0$ for monovalent metals such as Au [3,4,6] or Ag [7]. On the other side, magnetic materials have not yet been studied in detail and the possible lift of spin degeneracy in magnetic NW's represents still an open question [2,8].

Here, we have analyzed the room temperature electronic transport properties of atom-size metallic wires made of magnetic and nonmagnetic metals using an ultrahigh-vacuum (UHV, pressure $\leq 10^{-8}$ Pa) mechanically controllable break junction system (MCBJ) [2,6,7], which is a well established technique to study the conductance of nanostructures. In this approach, a macroscopic wire is glued on a flexible substrate in two points; then it is rendered fragile between the two fixing parts by an incomplete cut. By bending the substrate *in situ* in UHV, we break the wire and produce two clean metal surfaces. Using the same bending movement, the fresh tips are put together and separated repeatedly to generate and elongate NW's. It must be emphasized that the extreme cleanness of the environment and the sample itself are essential to get reliable and reproducible conductance data from atomic-size contacts [6,7].

In a MCBJ, the electrical transport of the metal NW's is measured using a two-point configuration; this implies that the conductance measurement probes the NW itself (the narrowest region of the contact) and the two leads (apexes). Although, the NW should show conductance quantization ($G = \alpha G_0$, where α is an integer), the electron reservoir-apex-NW coupling may act as an additional serial resistance, diminishing the conductance. Experimentally, α is frequently close to integer values but slightly lower.

In these experiments, an additional difficulty arises from the fact that a variety of conductance evolutions is observed. In fact, the structure of the NW or relative crystallographic orientation of the apexes cannot be controlled; then each conductance measurement corresponds to a new NW with a different atomic arrangement. To overcome this difficulty, most NW studies rely on the analysis of average behaviors of many conductance curves. The most frequently used procedure consists in building histograms from each individual electrical transport measurement, where occurrence of each conductance value is plotted (in this way a conductance plateau becomes a histogram peak). Subsequently, the so-called global histogram is constructed by the linear addition of individual histograms from a series of measurements. The presence of peaks close to integer multiples of the conductance quantum has been considered as the proof of quantized conductance in metal NW's [2].

The atomic structure of NW's generated by mechanical stretching has been studied using independent experiments based on time-resolved high resolution transmission electron microscopy (HRTEM), where the NW's were generated and elongated inside the microscope following the method reported by Kondo and Takayanagi [9]. NW's are generated by the following procedure: the microscope electron beam is increased to a current density of ~ 120 A/cm^2 and focused on a self-supported

metal film to perforate and grow neighboring holes. When two holes are very close, a nanometric bridge is formed between them. When these bridges are very thin (1–2 nm) and close to rupture, the electron beam intensity is reduced to its conventional value (10–30 A/cm^2) in order to perform the real-time imaging with atomic resolution. When the beam current is too high (as during the hole generation step), the film vibrates and no atomic imaging can be performed. The procedure described above has allowed us to generate NW's with a remarkable stability. In fact, the NW, its apexes, and the surrounding regions are all parts of a unique metal film and form a monolithic block. NW's formed by a few atomic layers usually show a long lifetime in the range of minutes. Although there is stability, the NW's elongate spontaneously, get thinner, and then break due to the relative slow movement of the NW apexes. This apex displacement is probably due to a film deformation induced by thermal gradients or just by low frequency vibration of the thin metallic film membrane, as usually observed in TEM thin film work [6,7].

The metal films (thickness 15 nm for Co; 5–6 nm for Pd and Pt) have been obtained by thermal or electron beam evaporation of pure metal on a substrate (usually NaCl crystals or freshly cleaved mica). Subsequently, the films are detached from the substrate by floating them in water; next, the sample is collected on a TEM holey carbon grid, remaining self-supported on the regions hanging over the holes. All images were acquired close to Scherzer defocus [10] using a high sensitivity TV camera (Gatan 622SC, 30 frames/s) associated with a conventional video recorder. The images were obtained by digitizing the video film *a posteriori*; in order to enhance signal-to-noise ratio, several frames (3–5) are usually added. This procedure has shown to be very efficient and has enabled us to register the NW real-time formation, evolution, and rupture, even for low atomic number metals as, for example, Co.

The steps mentioned above constitute the basic procedure for *in situ* NW observation. However, we must emphasize that the study of NW's has demonstrated to be a new challenge for each metal. Each material has required a particular setting, from the sample preparation (thin film thickness, etc.) to the NW generation and imaging. For this kind of experiment, noble metals seem to be a well-behaved case; on the other side, metals such as Co are more complicated, mainly because they are very reactive. We have prevented sample oxidation by evaporating sequentially carbon-metal-carbon layers on a substrate. This "metal sandwich" is detached from the substrate and collected on the TEM grid, as described above. Inside the TEM, the carbon layers are removed from a sample region using an intense electron irradiation, which can be very time-consuming (6–8 h).

Figure 1 shows typical behaviors of the conductance during elongation and thinning of Co NW's; also a histogram of occurrence of each conductance value (global histogram [2]) during a series of measurements is displayed in order to derive a statistical analysis of NW behavior. A quick analysis shows a major difference with previously studied metals (Au [2,6], Ag [2,7], Cu [2], Na [11], etc.): the last conductance plateau (or the lowest conductance peak of the global histogram) attributed to the thinnest Co wire is observed at $\sim 0.5G_0$ instead of $\sim 1G_0$.

In order to understand the origin of the $0.5G_0$ conductance, it is essential to determine the atomic arrangement of the thinnest possible Co NW. HRTEM imaging has shown that just before rupture, Co wires adopt a LCA configuration [see Fig. 2(a)], which must be associated to the last conductance plateau at $0.5G_0$. It must be noted that the global histogram, in Fig. 1(b), does not display a $1G_0$ peak, as could be expected by the addition of $0.5G_0$ steps. However, this is not surprising because it is already well understood that conductance curves are a signature of the structural evolution during the NW stretching [6]. Then, some conductance values may not be observed in a transport experiment, if there is not a stable

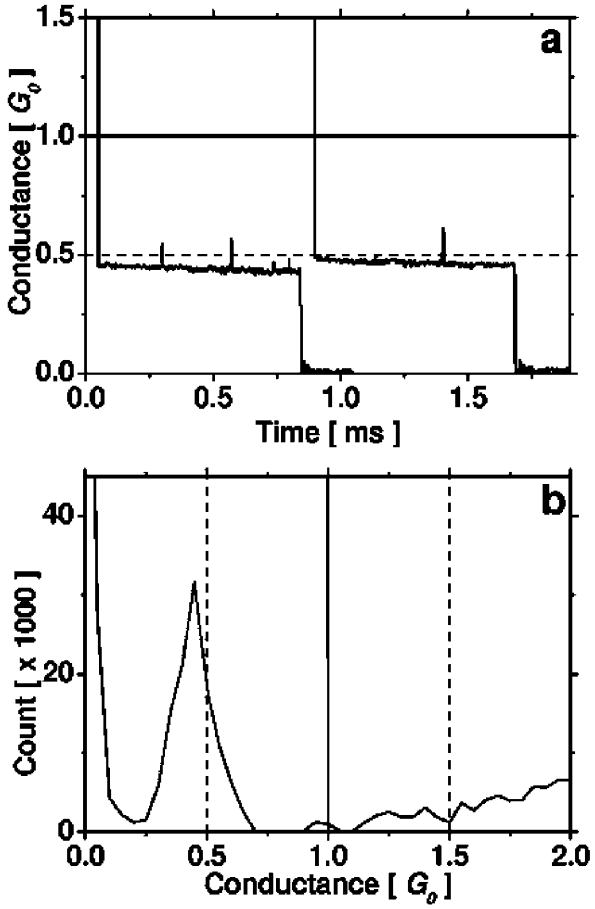

FIG. 1. UHV-MCBJ conductance measurements of Co NW's at room temperature and without external magnetic field. (a) Typical electrical transport curves showing conductance plateaus. (b) Global histogram exhibiting the statistical conductance behavior of a sequence of NW generations. Note that the plateaus in (a) and the lowest peak in (b) are located at $\sim 0.5G_0$

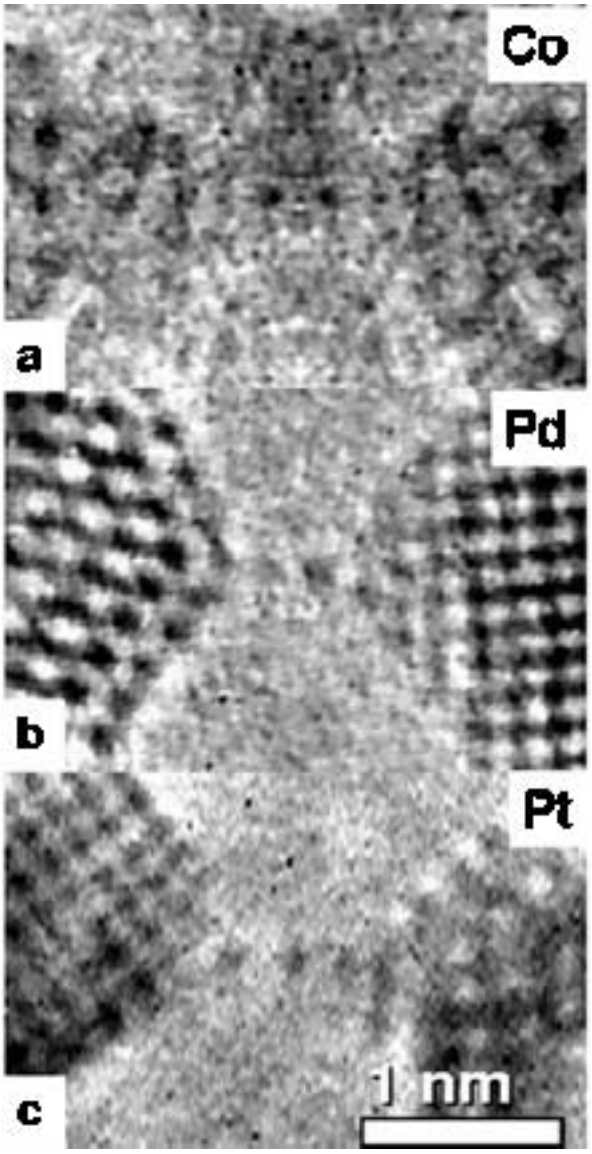

FIG. 2. HRTEM atomic resolved images showing the formation of suspended chains of atoms just before the contact rupture. (a) Co. (b) Pd. (c) Pt.

atomic structure sustaining that particular conductance value [7,12].

Conductance plateaus and jumps of $0.5G_0$ are observed for constrictions generated in bidimensional electron gazes when a strong magnetic field is used to lift the spin degeneracy; in fact, features at $0.5G_0$ are considered a signature of a spin polarized current generation [1,2,13–15]. Our results enable us to deduce that this phenomenon occurs spontaneously for one-atom-thick ferromagnetic wires in zero magnetic field and at room temperature. The polarized current can be originated either from a wide spin gap at the narrowest wire region or from the fact that the apexes contacting the atom chain have identical magnetic orientation (in other terms, magnetic domain walls may be expelled from the nanometric constriction). Further studies involving temperature or magnetic field dependence are in progress to elucidate this point. This behavior was observed not only in Co wires but also in NW's made of other ferromagnetic $3d$ transition metals such as Fe and Ni (not shown here). In contrast, it was not verified in similar experiments on Cu, a very close nonmagnetic $3d$ transition metal [2,16]. This kind of analysis can be applied only to the last conductance plateau ($0.5G_0$), and we are not able to derive further insights on the existence of polarized current in thicker NW's (or higher conductance), because the superposition of possible spin polarized and unpolarized conduction channels cannot be discriminated from the conductance value.

One-atom-thick wires represent the smallest achievable 1D system; then, they should be expected to show new and unexpected physical behaviors. For example, magnetic ordering may occur in low-dimensional systems of nonmagnetic materials [17]. It has been theoretically predicted that infinite 1D spin systems would not display magnetism [18]; nevertheless Gambardella *et al.* [19] have recently revealed magnetic ordering in chains of Co atoms deposed on Pt and spin blocks of 15 atoms at 45 K were reported. Certainly in this case, the Pt substrate must play a non-negligible role, but these results suggest that ferromagnetism could be expected in the short suspended chains analyzed in this work. On the other hand, it is surprising that the magnetic ordering seems to occur even at room temperature. In this sense, it will be very interesting to check if LCA's of quasimagnetic metals become magnetic and may show spin polarized conductance channels. The obvious test case would be the $4d$ transition metal Pd ([Kr] $4d^{10}$), where 2D thin films and 0D clusters have already been reported to become magnetic [20,21]. First, we have used HRTEM to show that the thinnest Pd nanowire displays a LCA structure [see Fig. 2(b)]. Subsequently, UHV-MCBJ experiments revealed that the global histogram shows the lowest conductance peak (associated to the LCA structure) at $0.5G_0$ [see Fig. 3(a)], as expected when a spin polarized current is allowed to travel through the Pd LCA's.

An extension of the previous studies to the $5d$ transition metals leads to Pt as a test metal candidate. For that row, Pt shows a stronger localization of the d wave function, although $5d$ metals have a very small exchange integral [22]. Concerning atomic structure, Pt NW's have already been shown to form LCA's [2,23]; see the example in Fig. 2(c). The results of conductance measurements of Pt NW's are shown in Fig. 3(b), where a well-defined peak at $0.5G_0$ can be easily recognized in the global histogram. The formation of a spin polarized current in Pt atom chains may seem surprising because the possible occurrence of magnetism in $5d$ transition metals has usually been neglected [22]. However, molecular beam photodetachment studies of very small (two to three atoms) Pt clusters have shown an anomalous behavior, somewhat following the tendency observed for magnetic clusters (Ni

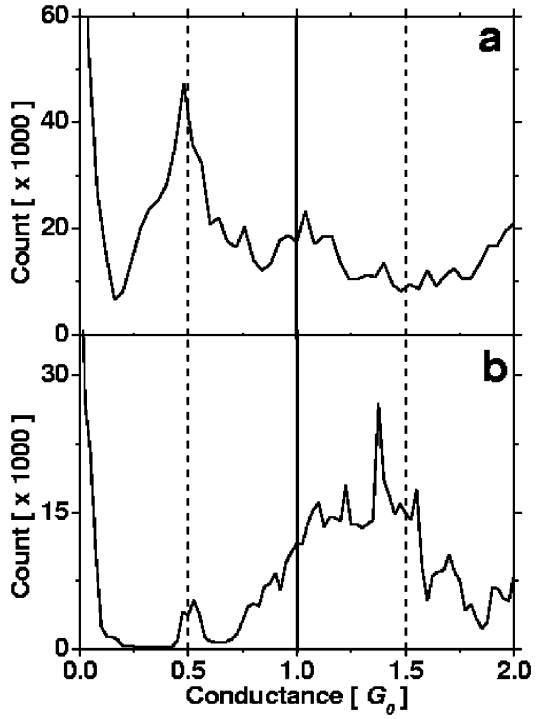

FIG. 3. Global histograms of conductance for (a) Pd and (b) Pt measured at room temperature and without external magnetic field. Note that the peak corresponding to the lowest conductance value is located at $\sim 0.5G_0$ for both metals.

or Pd) [20]. Also, we must keep in mind that in this size regime metal clusters adopt a linear structure [24], very close to the atomic arrangement in LCA's. In addition, recent *ab initio* calculations have predicted the generation of a magnetic state in highly elongated linear Pt chains [25,26]; the need of significant bond elongation could explain why the $0.5G_0$ peak is rather small for Pt, while dominant for Co and Pd.

In summary, we have shown that suspended chains of atoms made of ferromagnetic 3*d* transition metals display a conductance compatible with a fully polarized conduction channel ($0.5G_0$) at room temperature and without the need of external magnetic field. The 1D nature of LCA's also induces a similar conductance behavior in suspended chains of the quasimagnetic 4*d* metal Pd and a nonmagnetic 5*d* metal as Pt. These results open a wealth of new opportunities to get a deeper understanding of spin dynamics or spin control in nanostructures and will have important implications for the development of future spintronic devices. Although the practical application of a linear chain of atoms will be rather hard to achieve, their atomic structure provides a very good insight for designing and synthesizing new organometallic molecules containing magnetic atoms for applications in molecular electronics exploiting spin effects.

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