Huge anisotropic magneto-resistance in iridium atomic chains

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Abstract

We analyze in this article the magneto-resistance ratio of finite and infinite iridium and platinum chains. Our calculations, that are based on a combination of non equilibrium Green function techniques and density functional theory, include a fully self-consistent treatment of non-collinear magnetism and of the spin-orbit interaction. They indicate that, in addition to having an extremely large magnetic anisotropy that may overcome the super-paramagnetic limit, infinite and also realistic finite-length iridium chains show sizeable anisotropic magneto-resistance ratios. We therefore propose iridium nanostructures as promising candidates for nanospintronic logic devices.

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1 Introduction

The ability to enhance and tailor at the same time the magnetic anisotropy and magneto-resistance of atomic-sized magnetic bits and junctions will determine whether nanospintronics will be a viable technology. Cobalt clusters and chains with magnetic anisotropy barriers of the order of 1 meV were fabricated a few years ago by deposition on substrates of 5d elements[1,2]. Furthermore, several recent theoretical predictions have pointed out that atomic clusters and chains made of 4d elements should have even higher magnetic anisotropy barriers than Co nanostructures[3–5]. In the case of 5d elements like platinum and iridium, calculations predict that the anisotropies of iridium and platinum atomic clusters and chains can be even larger than room-temperature thermal energies, so that these nanostructures can overcome the technologically relevant superparamagnetic limit[4,6,7].

Suspended chains of gold atoms connecting two gold electrodes were fabricated using the scanning tunneling microscope and mechanically controllable break junction (MCBJ) techniques[8,9], where a quantized conductance close to $G_0 = 2e^2/h$ was measured. Subsequent experiments found that platinum and iridium also form suspended chains, but that other elements like Ni, Co, Pd or Rh do not[10,11], unless doped with oxygen or other elements[12]. It was also demonstrated that longer atomic chains could be made by deposition on stepped surfaces[11], or by encapsulation in carbon nanotubes[13].

The reduced dimensionality of nanometric objects increases their tendency towards magnetism. Ugarte and coworkers proposed that their experimental conductance data for platinum chains could be explained provided that these were magnetic[14]. Subsequent theoretical work indeed found that 4d and 5d infinite linear chains of bulk-paramagnetic materials do become magnetic[15,16]. Very recently, several groups have found that the magnetic anisotropy energy (MAE) per atom of infinite zigzag platinum and iridium chains[6,7], and of linear platinum chains[7] show values as large as 50 meV to 100 meV when stretched, which correspond to equivalent temperatures of 500 K to 1000 K. This suggests that the blocking temperature $T_B$ of such nanostructures will attain a similar value and therefore they should overcome the superparamagnetic limit, so that their magnetization would be finite at room temperature.

We draw the attention in this article to iridium and platinum atomic structures as promising candidates for nanospintronics. We show that iridium atomic chains not only have huge magnetic anisotropies, but also meet the requirement of having a significant magneto-resistance ratio. The simulations presented here are based on density functional theory[17] as implemented in the SIESTA code[18], and include a fully self-consistent and spin non-collinear implementation of the spin-orbit interaction[19]. This spin-orbit implementation has been successfully used to compute the magnetic anisotropy of infinite Ir and Pt atomic chains[6] and of transition metal atomic clusters[4], in agreement with the results of other simulations[3–7]. The conductance calculations were carried out with our spin non-collinear code SMEAGOL[20], which includes the fully self-consistent implementation of the spin-orbit interaction referred above. We note that SMEAGOL has been shown to provide conductance data that compare rather accurately with the available experimental data on platinum chains[10,11,21]. The simulations have used the Local Density Approximation[22] and norm-conserving pseudopotentials[23]. The atomic parameters that were fed in the pseudopotential generator, as well as the basis set used are similar to those reported in references[4,6,21].

Figure 1 (c) shows a schematic view of the experimental setup of a MCBJ experiment, where at the last stages before breakage of a Pt or Ir strip, a short suspended atomic chain is formed. In these experiments, an electric current is applied to the circuit, and the conductance $G$ is measured many times as the chain elongates. The whole setup can also be understood as a magnetic junction that connects two paramagnetic electrodes. Recent work has shown that theory can reproduce accurately the experimental conductance data of finite-length platinum chains[21], but has not found significant differences between the paramagnetic and the magnetic cases[24].

We show the energy of linear platinum and iridium chains as a function of $d_z$ in Figs. 2(a) and 3(a). It is important to...
notice that these chains are metastable. Indeed, allowing for lateral displacements of the atoms yields the energy curves in Figs. 2(b) and 3(b), that only have minima for ladder or zigzag arrangements similar to those shown in Figs. 1(a) and (b). Furthermore, relaxation of the forces always finishes in zigzag or ladder chains, unless the atoms are constrained to lie in a linear array. Actually, ladders are energetically more stable than zigzag geometries. But the relaxation of the forces in a MCBJ geometry, like the one shown in Fig. 1(c) only leads to zigzag arrangements, that gradually straighten as the electrodes are pulled apart. Related to this effect, we note that infinite platinum (iridium) zigzag chains become linear only for distances $d_z$ longer than 2.55 Å (2.40 Å), since at these distances, the zigzag angles fall abruptly to zero[6]. These values of $d_z$ are marked by a dotted vertical line in the right panels of Figures 2 and 3. We will therefore only present results for the conductance of infinite zigzag and linear, as well as finite zigzag, chains in this article.

We have performed separate simulations for orientations of the atomic spins along the chain axis ($z$- or easy-axis), perpendicular to it, but still in the zigzag plane ($x$-axis) and perpendicular to the zigzag plane ($y$-axis), but the energies $E_{x,y,z}$ can not be discriminated in the energy scale used in Figs. 2(a,b) and 3(a,b). We have therefore computed[4] the energy differences $\Delta_{x,y} = E_z - E_{x,y}$ per atom, that we define as the MAEs, and plotted them in Figs. 2(c,d) and 3(c,d) for platinum and iridium respectively. Our definition implies that when both $\Delta_{x,y}$ are negative, the magnetization lies parallel to the easy axis. On the contrary, when $\Delta_{x,y}$ is positive, the magnetization is oriented perpendicular to the easy axis, and aligns either parallel to the $x$- or the $y$-axes, depending on whether $\Delta_x$ is larger or smaller than $\Delta_y$, respectively. Notice that our results for linear platinum chains (Fig. 2(c)) are similar to those presented in Ref. [7]. More interestingly, the anisotropy of zigzag platinum and iridium chains is $x$-axis for short $d_z$, but shifts to easy axis if the chains are elongated, with the MAE achieving values as large as 50-80 meV depending on the specific value of $d_z$. The third row in Figs. 2 and 3 shows the spin moment per atom as a function of $d_z$. Smogunov and coworkers[7] found that linear platinum chains become magnetized at a shorter elongation $d_z$ for spin lying in the axis of the chain than for spins oriented perpendicular to it. This fact led them to predict a colossal magnetic anisotropy for the window of elongations within the onset of both magnetizations. However, we note here that the magnetization of the more realistic zigzag chains is finite for almost the full range of elongations regardless of the spin orientation, and therefore that effect is not expected to be seen. Interestingly, we find that the magnetization of iridium chains decreases when the chain is pulled apart, in a window of elongations around the equilibrium $d_z$.

Since infinite Ir and Pt chains show large MAEs, an obvi-
Spins along z-axis
Spins along x-axis

in Figs. 2(g,h) and 3(g,h), again as a function of the distance $d_z$.

FIG. 3: (Color online) Results of the simulations of infinite iridium atomic spin moments are oriented in the y-z plane. Two of these appear for unrealistic short elongations $d_z$, but the third develops for distances slightly larger that the equilibrium $d_z$ and therefore could have the potential to be realized and measured experimentally. Iridium chains show only one of these windows, where $G_z$ vanishes, which occurs for elongations about the equilibrium $d_z$, where the magnetic anisotropy favors orientations of the spins along the x-axis. The MAE in this window of elongations is of the order of 25 meV. Interestingly, the vanishing conductance develops precisely for orientations of the spins along the x-axis.

Since the above finite magneto-resistance ratios originate from the different number of channels available for electron conduction close to the Fermi energy for the different spin orientations, we now study the band structure of infinite chains. Indeed, the number of conductance channels can be found by counting the number of bands crossing the Fermi energy. We note that each band moves up or down in energy at a different pace as the chain elongates. Furthermore, since the spin-orbit interaction couples spin and orbital degrees of freedom, these shifts are different for the different spin orientations. Since the band structure of non-conducting chains must show a mini-gap at the Fermi energy, we have plotted in Fig. 4 the band structure of a platinum chain at $d_z = 2.45 \, \text{Å}$ for paramagnetic as well as for spin-orbit simulations. We indeed find that when the atomic spin point in the y-axis there exists a small mini-gap $E_g = 25 \, \text{meV}$ at $k = 0.4 \pi/d_z$. This value allows us to estimate the minimum number of atoms that a finite-sized platinum chain must have to develop such a mini-gaps. Using the relationship $N \simeq h v_F/E_g$, where we estimate the Fermi velocity at this $k$-point as $h v_F \simeq 1000 \, \text{meV} \, d_z$, we find a critical length of about 40 atoms. We therefore expect that it will be difficult to measure a significant magneto-resistance ratio in platinum chains. We have indeed simulated 5-atoms long platinum chains contacted to (001) platinum electrodes, in a geometry similar to Fig. 1 (c) and found that the magneto-resistance ratio was essentially zero.

In contrast, Fig. 5 shows the band structures of iridium chains, obtained from paramagnetic and also for spin-orbit simulations for an elongation $d_z$ such that $G_z$ falls to zero. We note that the spin-orbit interaction generates much larger gaps for iridium than for platinum. For this particular elongation,
Spins along z-axis

Spins along x-axis

Spins along y-axis

FIG. 6: (Color online) Conductance as a function of $d_z$ of iridium chains containing 21 atoms.

FIG. 5: Band structure of a zigzag iridium chain with an elongation $d_z = 1.96 \text{ Å}$. From left to right the panels show the bands for a paramagnetic calculation, and for spin-orbit simulations whereby the spins are oriented along the $z$, $x$- and $y$-axes, respectively.

the gap when the spins point along the $x$-axis is positioned exactly at the Fermi energy and has a value $E_g \simeq 100$ meV at the $\Gamma$ point. Using the estimated Fermi velocity $h v_F \simeq 1500$ meV $d_z$, we predict that chains containing about 15 atoms should show large magneto-resistance ratios.

To confirm this, we have simulated 2- and 21-atoms long iridium chains in contact to (111) iridium electrodes terminated by a pyramid, as in Fig. 1 (c), for a number of elongations of the chain, which cover both the equilibrium distance and also stretched situations. We have found that the magneto-resistive ratio of the 2-atom chains was negligible for whatever of these elongation. In contrast, we have found that $G_x$ is indeed substantially smaller than $G_{y,z}$, as we show in Fig. 6, rendering a finite ratio $AMR_x$. We stress that our previous analysis of Figs. 3 and 5 indeed led us to expect that $G_x$ had to be smaller than $G_{y,z}$.

To summarize, we expect that short platinum chains should show a negligible magneto-resistance ratio until the number of atoms in the chain exceeds of order 40. Since such a length is very difficult to attain experimentally, we believe that platinum is not the optimum magneto-resistance material. We predict that a better candidate element is iridium. Short iridium chains are expected to display small but measurable magneto-resistance ratios. Longer chains could develop perfect magneto-resistance for certain elongations. These clear differences are experimentally accessible and therefore we anticipate that future experimental studies of iridium chains will prove to be extremely fruitful.

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