Spin-dependent quantum transport effects in Cu nanowires

LETTER TO THE EDITOR

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D M Gillingham, C Müller and J A C Bland

Cavendish Laboratory, University of Cambridge, Cambridge CB3 0HE, UK

E-mail: jacb1@phy.cam.ac.uk

Abstract
In this work we investigate quantum transport in Cu nanowires created by
bringing macroscopic Cu wires into and out of contact under an applied
magnetic field in air. Here we show that a 70% magneto-conductance effect can
be seen in a Cu nanowire in a field of 2 mT at room temperature. We propose
that this phenomenon is a consequence of spin filtering due to the adsorption of
atmospheric oxygen modifying the electronic band structure and introducing
spin-split conduction channels. This is a remarkable result since bulk Cu is
not magnetic and it may provide a new perspective in the quest for spintronic
devices.

The very rapidly developing field of spintronics [1] is based on the proposal to use the spin
of the electron instead of its electrical charge in future devices. The challenge is to achieve
efficient spin injection at room temperature and to conclusively demonstrate the manipulation
of electron spin in an all-electrical manner. Spintronics requires small structures to satisfy
the requirement that the spins act coherently and that the electrons travel ballistically [2, 3].
Furthermore, low-dimensional structures such as nanowires can have vastly different electronic
structure compared to that of the bulk material and this can, on a sufficiently small scale, be
expected to be highly significant in the operation of spintronic devices. Here we present the
observation of magnetic field-dependent quantum transport in Cu nanowires. A large magneto-
conductance effect arises, which we attribute to the formation of spin-split conduction states
in the nanowire.

A nanowire can be considered to be an atomic sized constriction between two electron
reservoirs. If such a nanowire is small enough, the electrical conduction (G) can be quantized
according to the Landauer equation [4–6]

$$G = G_0 \sum_{n\sigma} T_{n\sigma} \tag{1}$$

where $G_0 = e^2/h$ is the conductance quantum (e is the electronic charge, h is Planck's
constant), $T_{n\sigma}$ is a transmission coefficient for the nth channel and electron spin σ (which
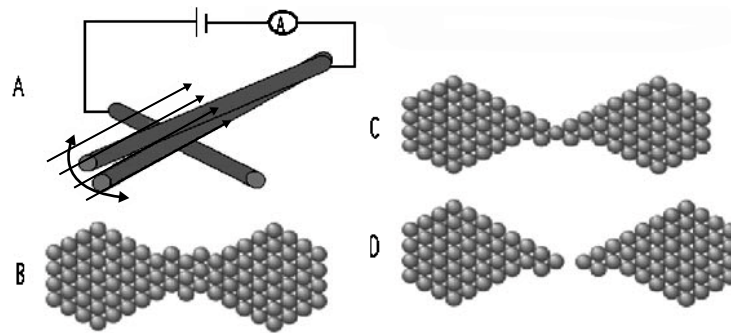
Figure 1. An outline of the experimental system used. (A) is a schematic diagram of the equipment; the nanowire is made where the wires come into contact. The magnetic field is applied in the direction of the dashed arrows. (B)–(D) give an illustration of how the nanowires are created. When the macroscopic wires are in contact, atoms bind to both wires (B); as the wires separate, the metal forms a neck which stretches out till it is atomic sized (C)—then it snaps (D). The quantum conduction is seen in region (C) close to (D) where the wires snap.

can take one of two values: either ↑ or ↓). For ballistic transport, $T_{n\sigma}$ can either be 1 or 0 corresponding to an open or closed channel. For non-magnetic materials one would expect the different spin channels to have the same energy and to be degenerate and in this case $G_0 = 2e^2/h$. It has been shown that for a two-dimensional electron gas [7] and ferromagnetic metals (such as Fe and Ni) [8–11] this spin degeneracy can be lifted. Garcia *et al* [12] have reported very large magneto-conductance effects of the order of 300% in Ni nanowires; however, Ni is a ferromagnetic metal and so magneto-conductance effects can be expected in this case.

The nanowires were made by tapping two macroscopic Cu wires together in air at room temperature [13, 14]. A schematic diagram of the experimental set-up is shown in figure 1(A); figures 1(B)–(D) outline the mechanism by which the nanowires are created. When macroscopic wires make contact with each other the atoms on the surface bond to atoms in the other wire (figure 1(B)). As the wires begin to separate, filaments are formed between the two wires (figure 1(C)). As the wires are separated further, the filaments are stretched and get thinner—in the nanoscale regime, as the wire gets thinner, conduction channels close and the conductance falls in steps. Towards the end of this process, there are only a very small number of atoms remaining in contact between the wires. At the end of the process the wires break (figure 1(D)). The reverse process can occur, giving rise to quantized conductance while the macroscopic wires are coming together.

The Cu wires have a 250 μm diameter, and purity of better than 99.99%, and were vibrated at a frequency in the range of 0.5–1 Hz. A bias voltage of 20 mV was connected across the two wires and the current flowing through the wires (about 1 μA per open conduction channel) was amplified by a transimpedance amplifier. Both the bias and the current were captured by a Tektronix TDS430A digital oscilloscope. Nanowires were not created every time the wires came together, so the data sets had to be filtered to separate those which demonstrated quantum conduction. A magnetic field was applied perpendicular to the nanowires via a pair of Helmholtz coils that were able to produce ±5 mT—as shown by the arrows in figure 1(A).

Figure 2 shows two typical conductance versus time curves obtained with no applied magnetic field; these illustrate two points. Firstly, quantum conductance can be seen in nanowires formed by the macroscopic wires: coming together, compressing the nanowires (figure 2(A)), and separating, stretching the nanowires (figure 2(B)). In general, the quantization
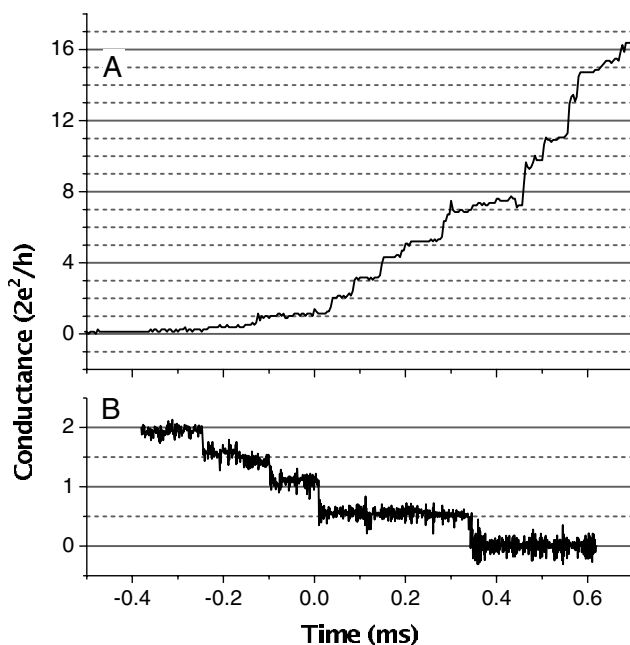
Figure 2. These are representative conductance versus time curves, taken from Cu at room temperature in air at zero applied field. The different signs of the curve gradients indicate that (A) was taken when the macroscopic Cu wires were coming together and (B) when they were separating. (A) shows the expected spin degenerate behaviour—i.e. the conductance quantized in units of $2e^2/h$; however, (B) shows a different behaviour: the conductance is quantized in units of e^2/h.

modes are the same for the nanowires formed by compression and tension, apart from the gradient of the curves; i.e. when the macroscopic wires are coming together the conductance starts low and ends high and vice versa when the macroscopic wires are separating. The second point relates to the size of the steps in the quantized conduction. Cu is a diamagnetic material, so from the Landauer theory presented earlier we expect the conductance quantum to be $G_0 = 2e^2/h$—that is, that the spin channels are degenerate. In figure 2(A) we see that the conductance quantum is indeed $2e^2/h$. However, when we inspect figure 2(B) we see a clear e^2/h quantization. These two conduction modes are seen whether the macroscopic wires are coming together or separating—so whether the nanowire is in compression or tension has no major effect on the mechanism causing the e^2/h quantization mode. From inspecting individual conductance versus time curves, it is apparent that the total collision time between the macroscopic wires was typically 5–50 ms.

Figure 3 shows two representative conductance histograms. These are built up by counting the number of points in conductance versus time curves which have a particular value of the conductance; thus stable conductance plateaux will show up as peaks in these histograms. So the peaks in the conduction histograms correspond to the values of the quantized conductance. Since each conductance versus time curve represents the creation of a nanowire, these histograms represent an average over many nanowires. Figure 3(A) was taken in zero applied field; as can be readily seen, peaks appear at odd multiples of e^2/h but they are small compared to the peaks at even multiples of e^2/h. Figure 3(B), however, was taken at a field of about 0.5 mT applied perpendicular to the nanowire (and parallel to one of the macroscopic
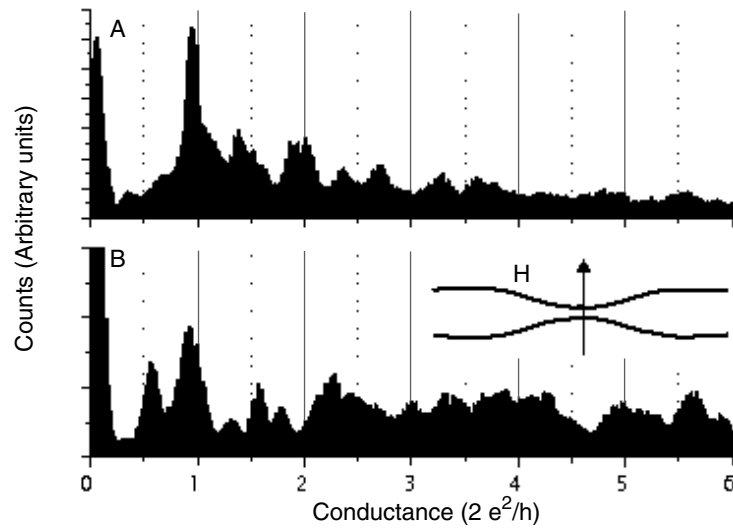
Figure 3. These are representative conduction histograms. They are built up from conductance versus time curves taken at room temperature in air. (A) was taken at zero applied magnetic field—it shows that the expected $2e^2/h$ quantization is dominant; however, there is a small peak at $0.5 \times 2e^2/h$ and one at $1.5 \times 2e^2/h$, showing that there is some quantization in units of e^2/h occurring. (B) was taken with a 0.5 mT magnetic field applied perpendicular to the nanowire—it shows that the e^2/h quantization has become far more dominant than in (A), the zero-field case. The inset is a schematic diagram of the field direction with respect to the nanowire.
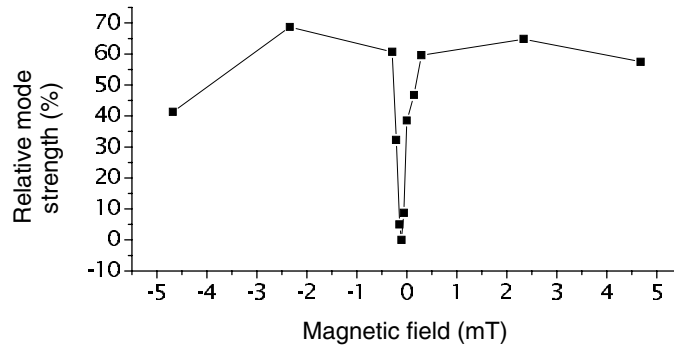

Figure 4. The magnetic field dependence of the relative strength of the e^2/h mode derived from the height of the e^2/h peak from conductance histograms normalized by the number of curves in the conductance histogram. The maximum is 70% at 2 mT. All measurements were made at room temperature. The field was applied parallel to the Earth's magnetic field and was perpendicular to the nanowire and parallel to one of the macroscopic wires—as shown in figures 1 and 3.

wires) as shown in the inset of figure 3(B). The e^2/h peak is now comparable in strength with the $2e^2/h$ peak.

Figure 4 shows the magnetic field dependence of the relative strength of the $G = e^2/h$ mode, which is defined as

$$\text{MC} = \frac{N\left(\frac{e^2}{h}, H\right) - N\left(\frac{e^2}{h}, 0\right)}{N\left(\frac{e^2}{h}, 0\right)}. \tag{2}$$

In figure 4, $N(e^2/h, H)$ corresponds to the height of the e^2/h peak at a magnetic field H obtained from a conductance histogram normalized to the number of curves that made up the histogram. This value corresponds to the stability of the contacts which give rise to this conductance value. We find a 70% magneto-conductance effect for $H = 2$ mT. Bulk Cu is a diamagnet, so this result is unexpected and also surprisingly large. For a conductance of e^2/h, a single conduction channel will be open; this is widely accepted as corresponding to the smallest possible contacts—generally a single atom (or chain of atoms). In our work the two quantization modes were both seen, with $G_0 = e^2/h$ and $2e^2/h$, but most importantly the relative strengths of these modes were observed to change with the applied magnetic field.

We next need to consider possible physical mechanisms which explain our principal findings, namely that the conductance quantum is either $G_0 = e^2/h$ or $2e^2/h$ and that a magnetic field dependence clearly exists. The possible mechanisms that we shall consider are based upon either (i) electron scattering due to disorder or impurities in the nanowire or (ii) the creation of a spin-polarized density of states (DOS) in the nanowire.

Structural disorder can cause elastic scattering of electrons to occur in the nanowire. Brandbyge *et al* [15] found that corrugated edges and localized scatters moved the peaks by an arbitrary amount—and so cannot cause the fixed e^2/h mode. de Heer *et al* [16] found that elastic scatterers can split the nanowire into two nanowires in series—so the conductance will be reduced. However, to obtain the e^2/h mode the two nanowires created by this method would need to have the same number of open conduction channels and it is not clear why this situation should arise. These mechanisms also do not explain the magnetic field dependence, although it has been shown that the magnetic field can increase the disorder in Au nanowires [17]: in that case a magnetic field strength of 10 T was required which is much larger than that used in this work.

Spontaneous magnetization of nanowires in a range of simple non-magnetic metals (Na, Cs and Al [18]) has been predicted. At critical radii these three-dimensional nanowires are predicted to undergo a transition to a spin-polarized magnetic state. Cu, like Na and Cs, is characterized by s electrons at the Fermi energy, so a spontaneous magnetization could arise within the Cu nanowire over a sufficiently large region to be thermally stable. Such a spontaneous magnetization will polarize the conduction electrons, and when an external magnetic field, sufficient to align the spin-polarized region in the direction of the field, is applied, it will lift the spin degeneracy between the spin-up and spin-down conduction channels; consequently the conduction mode will change from $2e^2/h$ to e^2/h. Similarly, an applied field is required for the observation of the e^2/h quantized conductance mode for nanowires made from bulk ferromagnets [10]. However, to our knowledge no conclusive experimental results on spontaneous magnetization formation in nanowires have been reported to date.

It is well known that oxygen can strongly modify the spin-polarized band structure in a wide range of materials (e.g. half-metallic behaviour arises in the case of some metallic oxides) and that adsorbates can affect the mechanical and electrical properties of nanowires [19]. Since this work was performed in air, the macroscopic Cu wires will inevitably have adsorbed oxygen. The adsorbed oxygen atoms can form part of the nanowires—and may become polarized when a magnetic field is applied. Oxygen can develop a spin-split p band at the Fermi energy, which has the effect of blocking one of the spin conduction channels and gives rise to the observed magneto-conductance effect. Absorbed oxygen is also one of the possible explanations for the large magneto-conductance effect seen in electrodeposited Ni nanowires [20]. In our work the magneto-conductance effect has been studied in the region when the nanowire will be at its thinnest and so single adsorbed impurities such as oxygen can easily play a role and give rise to the effect that we observe. The macroscopic Cu wires used were of high purity (the highest content of ferromagnetic impurity is that of Fe, at 1 ppm). This means that unless some

surface segregation effect concentrates the magnetic impurities at the surface, it is unlikely that magnetic atoms will take part in forming sufficient nanowires to cause the observed effect, whether by spin filtering [21] or by any other mechanism. In our view the oxygen adsorbate mechanism is the most likely.

To summarize, we have seen a significant magneto-conductance effect in Cu nanowires in fields of 2 mT. We interpret this as being due to the spin filtering effect introduced by oxygen adsorbates modifying the electronic band structure in the nanowire. In this work, surprisingly large spin-polarized quantum conduction effects have been seen in a material which in the bulk is non-magnetic, by reducing the dimensions of the active material: this may offer a new approach in developing spintronic devices, differing from the current research effort devoted to developing bulk materials which have 100% spin polarization. We conjecture that this effect could be seen in other non-magnetic metals when formed into nanowires under the correct conditions.

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