FROM A SINGLE-WALL CARBON NANOTUBE TO THE NANO-SQUID: A PRELIMINARY STEP TOWARDS MEASURING MAGNETIC PROPERTIES AT NANO-SCALE

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Abstract

We recently reported the fabrication of a nanosized Superconducting QUantum Interference Device (nano-SQUID) whose sensing heart-element was a single single-wall carbon nanotube (SWNT) as a part of the SQUID loop. Here, we will describe the various procedures involved in the making of this nano-device, then we will present some of the unprecedented transport phenomena related to their superconducting behaviour that was induced thanks to superconducting palladium/aluminium contacts, in the presence or absence of a mild magnetic field.

Introduction

The superconducting quantum interference device (SQUID) is a well-known and unique tool to study magnetic properties of solids. The main part of a SQUID (Figure 1)is a superconducting loop operating at liquid helium temperature, each branch of the loop being interrupted by a tunnel barrier, thereby making so-called Josephson junctions (Josephson, 1962).

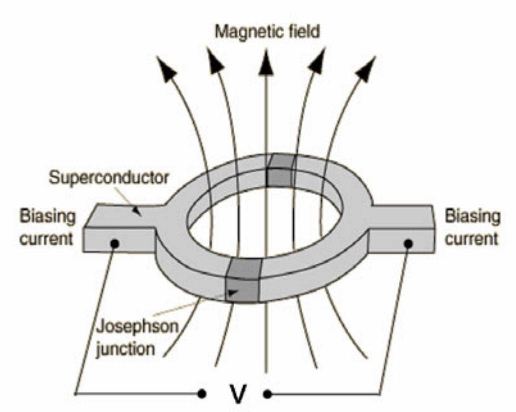

Figure 1. Principle of the SQUID sensing element.

A current is flown through the loop while subjected to a magnetic field. It divides in each branch and the electronic waves associated to each sub-currents are able to interfere once the two branches merge, with one period of voltage variation corresponding to an increase of one flux quantum. If another magnetic field is added, typically via the solid to measure, the two sub-current features re no longer identical giving rise to modifications in the interference figure. The sensitivity of the SQUID regarding the magnetic field threshold able to be detected is therefore related to the intensity of the smallest current detectable, which means that the superconducting loop has to be decreased in size as most as possible, down to nanosize. To overcome the difficulty to make nanosized tunnel barriers with the regular superconducting/insulating/superconducting sandwich structure, one way is to create a nanosized bottle neck within the same superconducting material, thereby creating a "weak link", according to the definition by Likharev (1979). That is where the aspect ratio and transport properties of single wall carbon nanotubes (SWNTs) made it possible.

Making the SWNT-Based SQUID (Nano-SQUID)

The main operation was to build two superconducting nanotube-based transistors using the same SWNT. We started from a degenerately n-doped silicon wafer substrate with a 350-nm-thick thermally grown SiO_2 layer on top, which was used as a backgate. The silica surface was first functionalized using a standard silanization technique, leading to a self-assembled monolayer of aminopropyltriethoxysilane. These substrates were periodically decorated with a specific pattern of converging gold micro-paths by means of optical lithography (Figure 2). Meanwhile, raw (i.e., not purified) SWNTs prepared by laser vaporisation and obtained from Rice University were dispersed under sonication added with sodium dodecyl sulphate (SDS) as surfactant, resulting in a highly diluted SWNT suspension. A micro-droplet of the resulting suspension was deposited onto the Si wafer at each of the areas where the gold micro-paths converge using a molecular combing technique (Gerdes, 1999) which includes 5 min dipping of the substrate into the SWNT suspension followed by withdrawing at 200 μm/s. This method allows a good control of the SWNT density and of their orientation on the substrate.

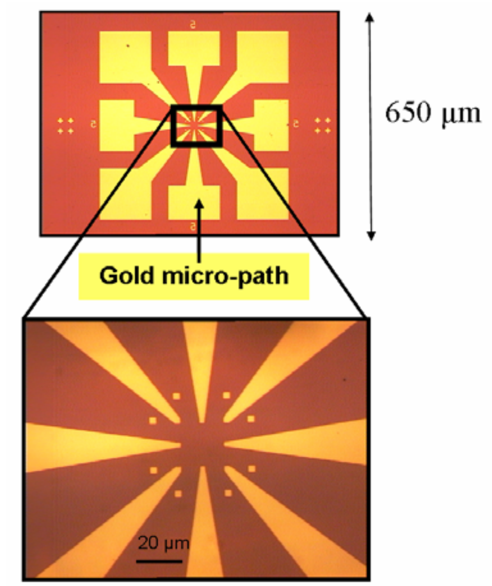

Figure 2. Sketch of the gold micro-path pattern deposited via optical lithography onto the doped Si wafers.

The sample was thoroughly washed in distilled water in order to remove the surfactant from the SWNTs. SWNTs suitably displayed onto the substrate were imaged by atomic force microscopy (AFM). Three bi-metallic electrodes (3 nm Pd followed by 50 nm Al) with a fork geometry were deposited on each of the selected SWNTs using aligned electron-beam lithography, so that the ensemble mimics the SQUID loop (Figure 3). Pd provides high-transparency contacts to the SWNTs (Javey, 2003), and Al is a superconductor with a critical temperature of about 1.2 K.

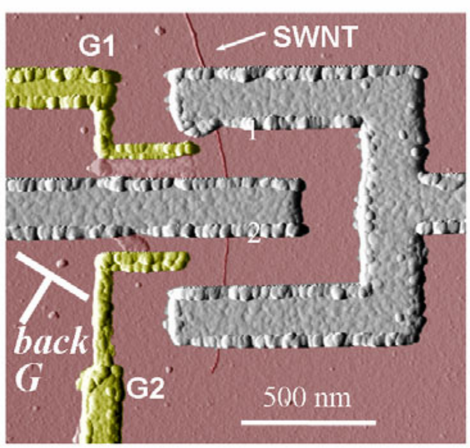

Figure 3. AFM image of a typical Nano-SQUID device. The three bimetallic contacts with the fork geometry make the SQUID loop (grey colour), in which the two uncoated portions of the SWNT (diameter ~1 nm) between the contacts act as the two Josephson junctions (with length of about 200 nm) of the loop. In addition, Two lateral gates G1 and G2 (gold colour) are added in front of the Josephson junctions, in addition to the back gate which is the doped Si substrate itself.

In addition to the back gate, two lateral gates G1 and G2 were aligned to each device, thereby providing each of the concerned portions of the loop the same configuration as a superconducting transistor. This allowed tuning independently the electronic properties of each SWNT junction (Fig. 1a). About 100 Nano-SQUID devices were prepared, among which about

30% were operational (at 35 mK). 70% rejection was caused by misalignments during superconducting path deposition, low conductance nanotube–metal contacts, and the occurrence of semiconducting SWNTs. Only devices with resistance below 30 kV and no significant gate effect at room temperature were kept for our studies.

Operating the Nano-SQUID

The operation of the CNT-SQUID is based on the quantum phase interference of the supercurrent flowing through two CNT-based superconducting transistors13 in a superconducting ring (Figure 4). The position of the quantum levels in each junction can be individually tuned using the two lateral gate voltages V_{G1} and V_{G2}, and the transparency of the CNT contact barriers can be globally adjusted using the backgate voltage V_{BG}. This provides an interesting facility where none, one out of two, or both junctions may be turned on or off (Figure 4).

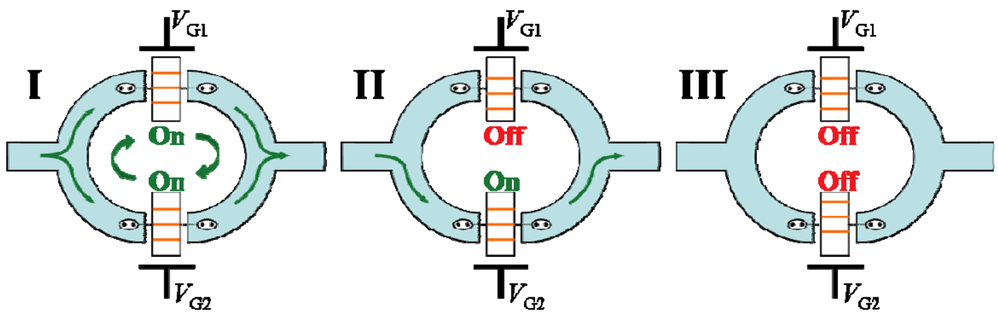

Figure 4. Sketches of the Nano-SQUID with two nanotube junctions, which can be tuned with the gate-voltages V_{G1} and V_{G2}. In case I, both junctions have a quantum level adjusted to the Fermi energy of the leads (on-resonance) and maximal supercurrent can flow through the device. In cases II and III, one and two junctions are tuned off-resonance, respectively.

Such a device provides an interesting tool for studying the fundamentals of electron transport in metallic SWNTs, whose detailed study was reported elsewhere (Cleuziou, 2006). Playing with the various gates and adding an outer magnetic field with various intensities allow obtaining a complete mapping of the electronic conductivity within the loop whose Figure 5 provides one example in function of the gate voltages.

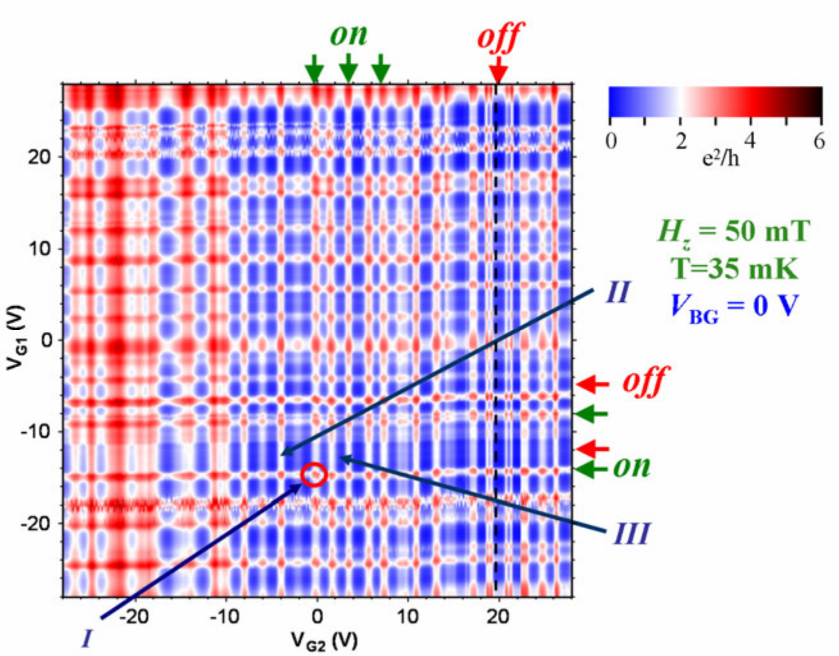

Figure 5. Correlation between normal-state conductance and superconducting switching current. Colour-scale representation of a typical differential conductivity dI/dV map at 35 mK and a backgate voltage V_{BG} = 0 as a function of the lateral gate voltages V_{G1} and V_{G2}. A magnetic field of H_z = 50 mT was applied perpendicular to the SQUID plane in order to suppress the superconductivity of the leads. The effect of cross-capacitance was subtracted in situ.

In this example, some of the on (green colour) or off (red colour) positions for each of the junctions are pointed out as well examples of the resulting current passing through the loop for on-on, on-off, and off-off configurations (corresponding to arrowed examples I, II, and III respectively). Current is maximum in red areas (up to four times the quantum of conductance) while it is minimum in blue areas. Other phenomena were revealed, e.g. related to weak or strong Kondo effects (Cleuziou, 2006).

In designing the Nano-SQUID, our motivation was to use it as a detector for magnetization switching of the magnetic moment of a single molecule. The aspect ratio of SWNTs makes them ideal for coupling to single nanometre-sized objects. In the case of a SWNT junction, a nanometre-sized molecule could be placed directly onto the SWNT, which has a cross-section of about 1 nm^2. A nearly optimized coupling factor α is therefore expected, because the molecule size and the junction cross-section are in the same range. Considering the Mn_{12} molecule as an example for molecular magnets, a rough estimate of the related magnetic signal yields a flux variation of 10^{-4} flux quantum, which is one order of magnitude higher than the estimated flux sensitivity of the device (Cleuziou, 2006). This means that measuring the magnetic flux from a single molecule should be possible. Another important feature of the Nano-SQUID concerns the ability to tune the coupling between the sensor and the molecule to investigate. Indeed, the supercurrent through the junction can be switched on and off using the lateral gate. In the off state, the magnetic molecule is decoupled from the measuring device and it can evolve without decoherence coming from the device. In order to measure the magnetization state of the molecule, the SQUID is then switched on. This should have important consequences as it should allow limiting the back-action of the Nano-SQUID on the quantum state of a single molecule magnet.

Conclusion

The SWNT-based SQUID (Nano-SQUID) provides a new generation of ultrasensitive magnetometers for nanosized-samples. Such devices also offer the opportunity to test interesting physical phenomena ranging from Kondo physics to p-junctions, and pave the way for non-locality experiments by generating pairs of entangled electrons in a nanotube.

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