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Carbon nanotube based magnetic flux detector for molecular spintronics

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This work describes the study of a superconducting quantum interference device (SQUID) with single-walled carbon nanotube (CNT) Josephson junctions. Quantum confinement in each junction induces a discrete quantum dot (QD) energy level structure, which can be controlled with two lateral electrostatic gates. The gates are also used to directly tune the quantum phase interference of the Cooper pairs circulating in the SQUID ring. Optimal modulation of the switching current with magnetic flux is achieved when both QD junctions are in the 'on' or 'off' state. In particular, the SQUID design establishes that these CNT Josephson junctions can be used as gate-controlled π junctions. Besides, the CNT-SQUIDs are sensitive local magnetometers, which are very promising for the study of magnetization reversal of an individual magnetic particle or molecule placed on one of the two CNT Josephson junctions.

1 Introduction

The superconducting quantum interference device (SQUID) has been used very successfully for magnetometry and voltage or current measurements in the fields of medicine, metrology and other fields of research [1]. It combines two quantum properties of superconductivity: the tunnelling of Cooper pairs through a nonsuperconducting medium (the Josephson effect) [2] and flux quantization in a superconducting loop. The direct-current version of this device (DC-SQUID) is composed of a superconducting loop having two Josephson junctions. Its most striking property is that the maximum superconducting current flowing through the device can be periodically modulated by the magnetic flux entering the loop, with a period equal to the flux quantum. Recently, miniaturized versions of these devices have been used to implement phase qubits or to measure quantum magnetization reversal of nanoparticles [3] and single-molecule magnets [4].

We combine here the research area of SQUIDs with that of electronic transport through molecules in order to design a new detector: a SQUID with molecular Josephson junctions made out of CNTs [5]. This system allows us to address the problem of resonant tunnelling through a QD having discrete energy levels and coupled to superconducting electrodes. The SQUID has the advantage of linking the phase across both junctions with the magnetic flux entering the loop. It then yields insight into the current–phase relation across a QD coupled to superconductors. In particular, a gate-controlled transition from the normal to the π-junction can be observed [6, 7]. Owing to the geometrical aspects of CNTs, such SQUIDs are also very promising in the study of spin states of an individual magnetic molecule placed on one of the two CNT Josephson junctions.

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2 Results

2.1 SQUID design and CNT junction

We designed and built the first CNT-SQUID as presented in Fig. 1. The fabrication of the devices is described elsewhere [6]. The SQUID is composed of a superconducting loop, interrupted in two places by two carbon nanotube weak links. The fork geometry for the loop allowed us to fabricate both Josephson junctions from the same nanotube. Metal electrodes were composed of bilayer of Pd and Al. Pd provides high-transparency contacts to the CNTs [8]. Al is a superconductor widely used in nanoscale devices, having a critical temperature of about 1.2 K. In addition, two lateral gates G1 and G2 were aligned to the devices, allowing us to tune independently the electronic properties of each CNT junction.

The CNT-SQUID contains two CNT-based superconducting transistors, which have been described previously [9] and can be modelled by a QD between two superconducting leads (Fig. 1b). The position of the quantum levels can be tuned with gate voltages V_{G1} and V_{G2}. When a quantum level is aligned with respect to the Fermi energy of the superconducting leads (‘on’ state in Fig. 1b), a supercurrent of a few nanoamperes can flow by resonant tunnelling through the CNT. When the quantum levels are far from the Fermi energy (‘off’ state in Fig. 1b), the supercurrent is strongly reduced (typically by a factor of 10 to 10^3) [6].

2.2 Electronic transport properties and Kondo effect

Before turning our attention to the CNT-SQUIDs, we have to characterize the electronic transport properties of our CNT junctions. Particularly important is the interplay between Kondo correlations and superconductivity, which has recently motivated theoretical [10, 11] and experimental studies [12, 13]. Indeed, for strongly hybridized QD states, it has been theoretically predicted [11] that the Josephson coupling could be enhanced by the Kondo resonance. This effect partially offsets the reduction in Josephson

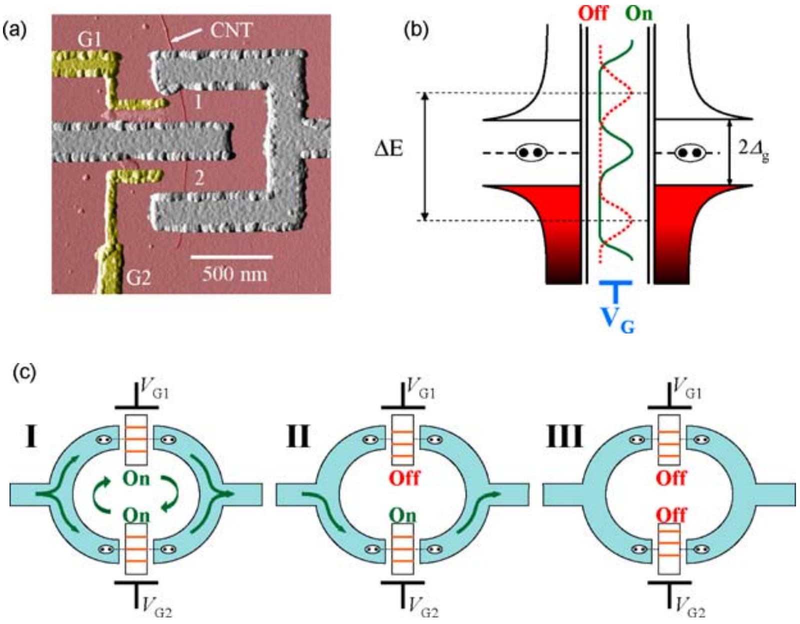

Fig. 1 (online colour at: www.pss-b.com) Device and operation scheme: (a) Atomic force microscope (AFM) image of the CNT-SQUID with two lateral gates G1 and G2. (b) Energy level schematics of a QD between two superconducting leads. The position of the quantum levels can be tuned with a gate voltage V_G. Only when a quantum level is adjusted to the Fermi energies of the superconducting leads (green curve) can a strong supercurrent flow between the leads. (c) Schematics of the CNT-SQUID with two nanotube junctions, which can be tuned with the gate voltages V_{G1} and V_{G2}.

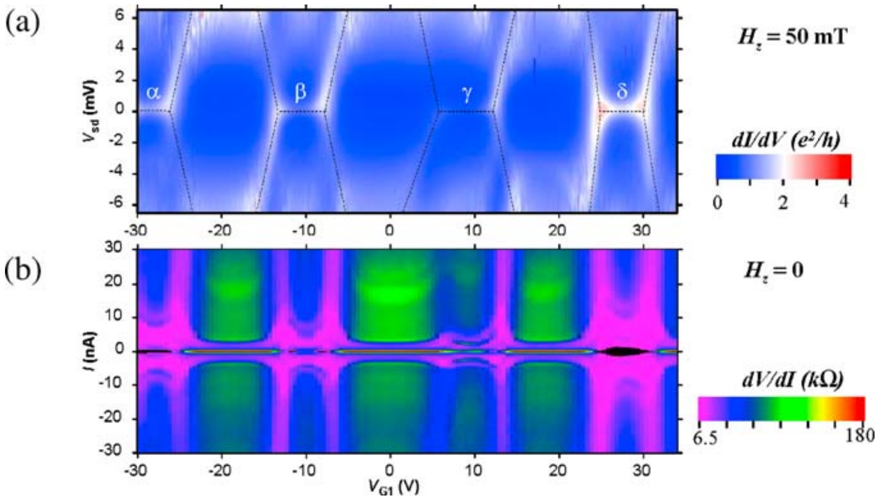

Fig. 2 (online colour at: www.pss-b.com) Correlation between Kondo effect and superconductivity: (a) dI/dV map as a function of V_{G1} and source–drain voltage V_{SD}. $V_{G2} = -6$ V and $H_z = 50$ mT. The dotted lines indicate the Coulomb diamonds, which are connected by Kondo ridges labelled α, β, γ, δ. (b) Differential resistivity dV/dI map of the same V_{G1} region and for $V_{G2} = -6$ V, but for $H_z = 0$. A supercurrent is observed in the black regions, which correspond to the Kondo ridges in (a).

coupling due to the Coulomb repulsion energy U_c. The Kondo resonances of each QD can be studied independently by measuring the differential conductance dI/dV as a function of source–drain voltage V_{SD} and lateral gate voltage V_G while keeping the chemical potential of the other QD at a constant value. The measured conductance map of dI/dV versus V_{SD} and V_G exhibits the typical features of Coulomb diamonds (Fig. 2), which are connected by Kondo 'ridges' of enhanced conductance. The enhanced conductance at zero bias results from the Kondo resonance when there are an odd number of electrons on the QD [14]. Note that the Kondo ridges appear at the same gate voltages as the occurrence of a superconducting state (Fig. 2b). Moreover the maximum supercurrent coincides with the most prominent Kondo ridge (labelled δ in Fig. 2a), a fact that supports the enhancement of superconductivity by the Kondo resonance [10, 11]. The tunnel rate $\Gamma \sim 1$ meV/h (where h is the Planck's constant), the Coulomb energy $U_c \sim 6$ meV and the energy-level spacing $\Delta E \sim 9$ meV were obtained from the size and shape of the Coulomb diamonds.

2.3 CNT-SQUID characteristics

The operation of the CNT-SQUID is based on the quantum phase interference of the supercurrent flowing through two CNT-based superconducting transistors [9] in a superconducting loop (Fig. 1c). 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} [15]. In order to find the gate voltages required to adjust the levels of each CNT junction with respect to the Fermi energy of their contacts, we measured the differential conductance dI/dV at zero bias as a function of V_{G1}, V_{G2} and V_{BG} in a field of $H_z = 50$ mT to drive the superconducting leads to the normal state (Fig. 3). The map follow the stability diagram of two QDs in parallel, that is, a chequerboard pattern with high-conductance states having values of the order of 4 e^2/h centred on points where both CNTs are on-resonance, and low-conductance states (typically between 0.1 and 0.5 e^2/h), where both CNTs are off-resonance.

At zero or small applied fields ($H_z \ll 50$ mT), the CNT junctions transmit a supercurrent. We measured the switching current I_{sw} map (maximal supercurrent) at the same gate voltage range. We observed at $H_z = 0$ that I_{sw} is maximal (about 6 nA) at maximal conductance in the normal state, that is, when both CNTs are on-resonance. I_{sw} is about two times smaller when only one CNT is on-resonance [6].

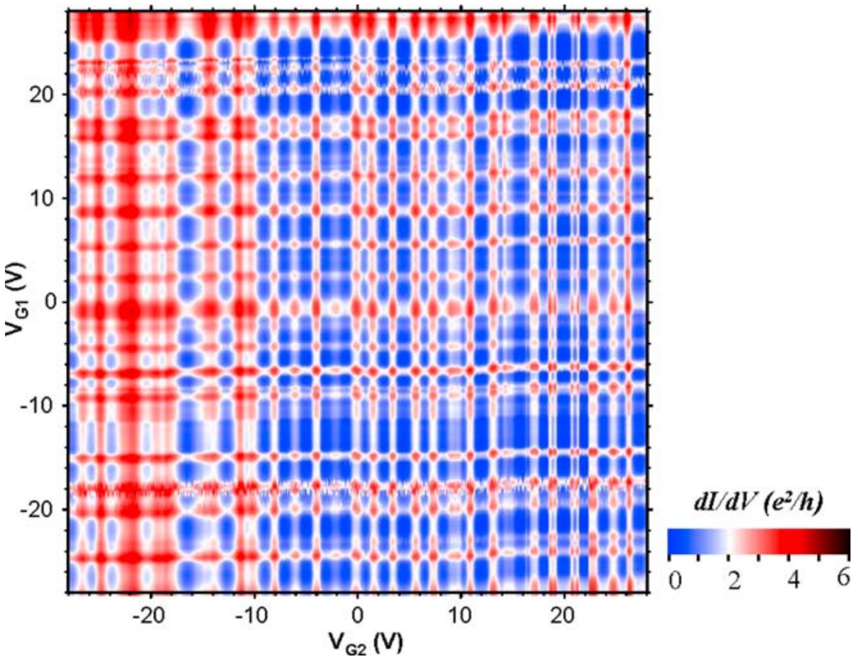

Fig. 3 (online colour at: www.pss-b.com) Normal state conductance: Colour-scale representation of a dI/dV map at 34 mK and $H_z = 0$ (normal state) as a function of the lateral gate voltages V_{G1} and V_{G2}, at a backgate voltage $V_{BG} = 0$ V. The effect of cross capacitance was substracted in-situ.

Provided the critical currents of each junctions are quite comparable, they can interfere in the SQUID loop. The maximal supercurrent oscillates as a function of the magnetic field H_z applied perpendicular to the SQUID (Fig. 4). The field periodicity times the area of the SQUID loop is in good agreement with a flux quantum ($h/2e$).

A closer investigation of the field modulation shows that, under certain gate voltages, the contrast of the interference fringes drops to zero and finally exhibits a phase shift, leading to a minimum switching current at zero field [6] (data not shown). For a weak Kondo effect, the large on-site interaction only allows the electrons in a Cooper pair to tunnel one by one via virtual processes in which the spin ordering of the Cooper pair is reverse, leading to a negative Josephson coupling (π-junction) [10].

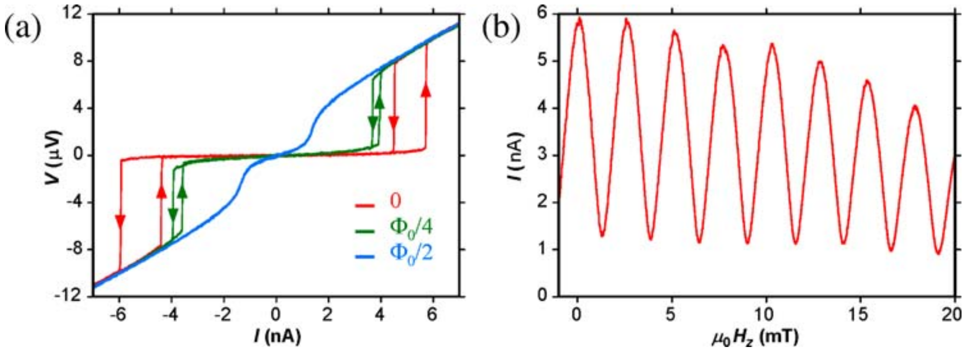

Fig. 4 (online colour at: www.pss-b.com) CNT-SQUID characteristics: (a) Voltage versus current curves at three applied fields corresponding to zero (red), a quarter (green) and a half (blue) flux quantum, when both junctions are tuned on-resonance. (b) Field modulation of the switching current I_{sw} for the situation in (a).

2.4 Prospects for detecting single magnetic moments

In designing the CNT-SQUID, our motivation is to use it as a detector for magnetization switching of the magnetic moment of a single molecule. Indeed, SQUIDs are the most sensitive magnetic flux detectors and the aspect ratio of CNTs makes them ideal for coupling to single nanometre-sized objects. A nanometre-sized molecule could be placed directly on the CNT, leading to an optimised flux coupling factor with the SQUID loop, since the size of the molecule is comparable to the CNT junction cross section. A rough estimate of the magnetic signal of a Mn_{12} molecule, sitting on the CNT, yields a flux variation of 10^{-4} flux quantum. When averaging I_{sw} during 1s at a rate of 10 kHz, we estimated a sensitivity of about 10^{-5} flux quantum (see Supporting Information [6] for the first estimation of the flux sensitivity of CNT-SQUIDs), which should be within the sensitivity of our measurements. The sensitivity estimation was preliminary and we expect to improve the sensitivity by measuring I_{sw} at faster rates, by using a new readout scheme [16], attractive for ultra-high sensitivity magnetometry, or by using the SQUID as a threshold detector [17] in the picoampere region. Using Al as a superconductor, the working temperature of our devices is limited for a few hundred millikelvin and further improvements could be achieved by using other superconducting materials with higher T_c, such as Nb, Pb, . . .

In order to depose magnetic molecules onto the CNT-SQUID, we are going to use and electrospray technique enable to depose the molecules in a vapour directed into the dilution refrigerator. Another possibility could be the functionalization of the CNT and the controlled attaching of molecules with adapted ligands.

3 Conclusion

In conclusion, the CNT-SQUIDs provide a new generation of ultrasensitive magnetometers of nanometre-sized samples. Such devices also offer the opportunity to test interesting physical phenomena ranging from Kondo physics to π-junctions.

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