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Hybrid organic spin valves

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Summary

Spin electronics - spintronics - is the field of electronics that uses the spin property of the charge carriers (electrons or holes) to transport and manipulate information. The simplest spin based device is the *spin valve* in which a non magnetic material is placed in between two ferromagnets. When a current is send trough, two resistance states are measured (low or high), depending on the relative orientation of the magnetization of the ferromagnetic electrodes (parallel or antiparallel). The ferromagnetic electrodes act as injector/detector of spins whereas the nonmagnetic material represents the transport medium for spins. For a spin valve device to function it is required that (at least a part of) the spins survive while being transported trough the non-magnetic material, that is, the non-magnet must be shorter than the *spin flip length* λ_{sf} (the length after which the spins have flipped). The spin flip length is proportional to $\sqrt{\mu\tau_{sf}}$, where μ is the mobility of the carriers and τ_{sf} is the spin relaxation time. Therefore, a high carrier mobility and a long spin relaxation time are needed in order to have a long spin flip length. In general, due to the low spin orbit interaction in light materials, the organic semiconductors (mostly made up of light carbon atoms) have a long spin relaxation time. Among the organic semiconductors, pentacene has one of the highest mobilities. These aspects make pentacene a good candidate for organic spin valves. For applications, devices which operate at room temperature are desirable, a reason why ferromagnetic metals are the best candidates since their Curie temperature is well above room temperature. Building Co and pentacene based spin valves is a subject of great interest from both fundamental and commercial point of view and constitutes the subject of this thesis.

The brief theory of spin valve devices presented in *Chapter 2* provides the motivation for the experiments presented in the subsequent chapters. In the frame of simple resistor model, it was shown that hybrid metal-semiconductor spin valves suffer from the conductivity mismatch problem: the low and spin dependent resistivity of the injector/detector with respect to the high and spin independent resistivity of the semiconductor prohibits efficient spin injection/detection. The solution to this problem is to properly match the conductivity of the injector/detector with the one of the semiconductor. An alternative to realize this is to insert tunnel barriers in between the ferromagnetic metal and the semiconductor. Since tunneling conserves the spin orientation, the resistance of the tunnel barriers is to be taken into account as an extra (spin dependent) resistance added to the injector/detector. Additionally, in a classical semiconductor picture, the existence of the Schotkky barrier at the metal/semiconductor interface may allow efficient injection or detection of spins. These theoretical aspects motivated us to study the interfacial properties of pentacene with Co in the clean contact case and upon the insertion of thin aluminum oxide (AlOx) tunnel barriers. The investigations are conducted using X-ray and ultraviolet photoelectron spectroscopies (XPS and UPS) which allow the study of the interfacial energetics (chemical interaction and energy level alignment). The results of these investigations constitute the core results of this thesis and are presented in *Chapter 4* and *Chapter 5*. Ultimately, spin valve devices were fabricated and characterized, results which are presented in *Chapter 6*. Experimental details regarding the fabrication of the samples and the experimental techniques used for their characterization are presented in *Chapter 3*.

The energy level alignment symmetry at Co/pentacene/Co interfaces was studied by means of XPS and UPS (*Chapter 4*). We found similar injection barriers of about 1.0 eV regardless of the fact that pentacene is deposited on Co or vice versa. The interfacial dipoles were found to be asymmetric, a characteristic which was attributed to the high roughness of the surface layer in the case of Co deposition on pentacene. The band diagram of a complete Co/pentacene/Co layered structure was drawn in the frame of a rigid band model with interfacial dipoles. Also, we were able to determine that mixing of pentacene orbitals with the ones of Co (hybridization) occurs and that Co atoms do not diffuse significantly when deposited on the loose matrix of pentacene. For spin valve devices and from the conductivity mismatch problem point of view, the existence of significant carrier injection barriers may allow efficient spin injection in pentacene under an applied bias when the barrier becomes triangular and direct tunneling from Co into highest occupied molecular orbital (HOMO) of pentacene is possible. However, to properly function a spin valve device must have both injector and detector resistances properly matched with the one of pentacene. In this respect, spin valve devices with clean contacts Co/pentacene is still problematic and the insertion of tunnel barriers appears as necessary.

In *Chapter 5* the energy level alignment scheme at Co/AlOx/pentacene was investigated as a function of the AlOx barrier thickness and the oxidation state of Co by means of XPS and UPS. The energy level alignment at Co/AlOx interfaces was found to be consistent with the formation of an interfacial dipole, its magnitude being sensitive to the oxidation of Co, and band bending in the thin AlOx tunnel barrier. The vacuum level of pentacene aligns with that of the Co/AlOx substrate in all cases, the injection barrier being determined solely by the Co/AlOx interfacial properties. The experiments showed no hybridization effects at the AlOx/pentacene interface. The hole injection barrier increases with thickness of the tunnel barrier and decreases with the oxidation of Co at fixed AlOx thickness. Interesting enough, the hole injection barrier for the thinnest tunnel barrier, which was derived from oxidizing 6 Å Al, was found to be about 0.4 eV smaller than the one found for the clean contact case. This means that devices with thin oxide tunnel barriers should perform better electrically. The variation of the hole injection barrier with the tunnel barrier thickness allows in principle the tuning of the spin injector/detector resistance and matching it with the pentacene resistance.

Pentacene based devices with Co, Au, Co/AlOx electrodes were fabricated and characterized, results which are presented in *Chapter 6*. The current voltage measurements of Co/pentacene clean contact devices were found to be (qualitatively) consistent with the photoelectron spectroscopy measurements. In lateral devices, due the sample fabrication procedure, Co oxidation which is detrimental to spin injection could not be avoided. In contrast, layered devices fabricated *all in situ* had the disadvantage of the ill defined (rough) top pentacene/Co interface, which caused uncontrolled magnetic properties of the top ferromagnetic electrode. Magnetoresitance measurements on layered clean contact devices were consistent with spin valve behavior, contrary to expectations. However, whether they truly represent spin valve behavior could not be unambiguously proved. In order to solve the conductivity mismatch problem a field effect geometry with AlOx tunnel barriers of a fixed thickness (the natural oxide thickness) was used. In this geometry, the pentacene resistance is the one that is fine tuned in order to match the fixed resistance of the tunnel barriers. No spin signal was observed. The noise in these measurements was of the order of few percent. Most likely, this hinders the measurement of the small spin signal.