NANO-STRUCTURES HYBRIDES SUPRACONDUCTEUR-FERROMAGNETIQUE

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Resumé. Le domaine des nano-structures hybrides Supraconducteur-Ferromagnétique (S/F) a rencontré un intérêt croissant dans ces dernières années. En effet, les structures hybrides permettent d'étudier l'influence mutuelle entre deux états fondamentaux de la matière condensée antagonistes : l'ordre ferromagnétique et la supraconductivité singulet. L'ordre ferromagnétique tend à aligner les spins des électrons pendant qu'une paire de Cooper est formée d'électrons de spin opposé. Pour des systèmes massifs, la coexistence entre ces deux états ordonnés demande des échelles d'énergie caractéristiques comparables, une condition difficile à réaliser expérimentalement car l'énergie caractéristique de la supraconductivité est l'énergie de formation d'une paire de Cooper ˜ 1meV pendant que l'énergie caractéristique de l'ordre magnétique est l'énergie d'échange typiquement ˜ 1 eV. L'originalité des structures hybrides est de s'affranchir de cette contrainte particulièrement lourde car le ferromagnétisme et la supraconductivité coexistent uniquement aux interfaces et se « parlent » par les conditions aux limites. Cette interaction « de voisinage » se révèle particulièrement efficace. Malheureusement l'antagonisme entre l'ordre ferromagnétique et la supraconductivité réduit l'échelle de coexistence à quelque nanomètre à l'interface entre les deux systèmes. La contrainte sur les énergies caractéristiques propres des systèmes massifs à été remplacée ainsi par la contrainte d'échelle dans des heterostructures S/F. C'est donc uniquement grâce aux développements des techniques de préparation d'échantillons de dimensions réduites que des expériences nouvelles ont pu voir le jour. Ce cours veut présenter les effets de l'ordre magnétique sur le condensat superfluide. Il est divisé en deux parties une première sur les propriétés d'équilibre et les perspectives d'expériences originales de Mécanique Quantique Macroscopique, une deuxième sur l'injection de spin polarisé et ses applications potentielles dans des dispositifs à trois terminaux. Cette note est une introduction au cours.

Proximity effect and Inhomogeneous Superconductivity.

In a BCS superconductor Cooper pairs form below the critical temperature, T_c. They are made of two electrons of opposite spin and Fermi momenta. At the same temperature a macroscopic number of Cooper pairs condense in a superfluid. The condensate is described by a macroscopic wave function. This wave function is a constant in homogeneous superconductors at the equilibrium and it defines the number of Cooper pairs. How the condensate wave function is modified by the exchange field in a ferromagnetic superconductor ? Do the Cooper pairs survive to the Zeeman splitting ? Actually, the superconducting state disappears when the exchange energy is larger than the paring energy, i.e. the superconducting energy gap, Δ_S. Because the energy gains by the Cooper pair condensation is lower than the energy looses to maintain the Cooper pair in the singlet state. Thus Clogston [1] showed that in a 3D superconductor the normal state is recovered when $E_{ex}>?\sqrt{2}/2\Delta_S$. However, Fulde and Ferrel [2], and Larkin and Ovchinnikov [3] (FFLO), showed independently that a new inhomogeneous superconducting state can show up nearby the Clogston limit. Unfortunately this state occupies a tiny part of the superconducting phase diagram and it is very sensitive to atomic disorder. The main reason is that the self-consistent gap equation requires for this inhomogeneous superconducting state to occur not only that the Clogston criterion is satisfied but also that the superconductor is in the clean limit. Despite a large amount of work in ternary rare-earth compounds [4], Chevrel phases [5], and the new found borocarbites [6] and rutheno-cuprates [7] [8], a microscopic experimental evidence for this state is still missing.

The situation is more favorable if Cooper pairs are injected from a superconductor into a ferromagnet, F, by the proximity effect. Assuming that the exchange field weakly affects the superconductor, superconducting correlations persist in F even for exchange energies much higher than Δ_S. The physical reason is that Cooper pairs are not instantaneously broken when they penetrate into the ferromagnet. They survive for a time corresponding to a traveled length on the order of $\xi_F=\hbar v_F/2E_{ex}=1/Q$, the coherence length scale in F [9], which is independent of the energy gap. The breakdown of the Clogston criterion turns out to be very significant since E_{ex} is usually much larger than Δ_S. Furthermore, energy conservation requires that a Cooper pair entering into a ferromagnet receives a finite momentum, Q, from the spin splitting of the up and down bands. This is illustrated in fig.1a. By quantum mechanics, Q modifies the phase, $\varphi=Qx_F$ of the pair wave function that increases linearly with the distance, x_F, from the S/F interface [9]. If all pair trajectories and spin configurations are taken into account, the amplitude of the superconducting order parameter in F is given by $\sin(\varphi)/\varphi$ (see fig.1b) [10]. Therefore, the superconducting order parameter induced into F oscillates around zero as a function of the distance from the S/F interface. The oscillations length is the same as the decay length and it is given by ξ_F. As only phase coherence is required in F, this state, contrarily to the FFLO state, is not sensitive to elastic scattering. Of course, the phase is accumulated by the Cooper pairs in F provided the electron spin is a good quantum number. Spin-orbit interaction reduces the spin mean free path, l_s, and the oscillations disappear for $l_s<\xi_F$ [9].

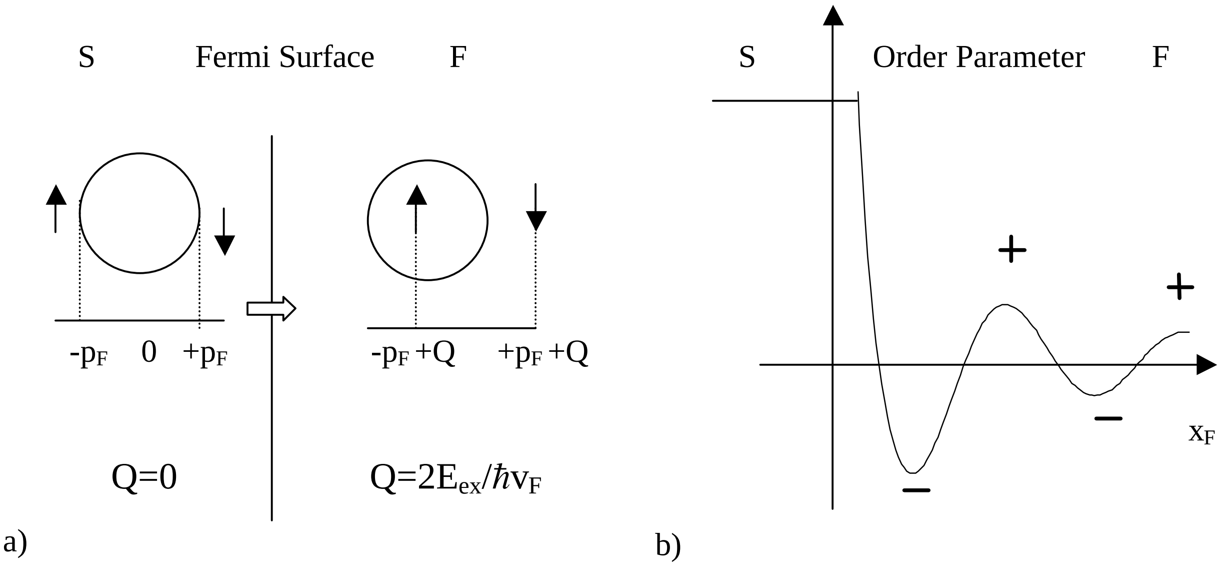

Fig.1 : a) A Cooper pair receives a finite momentum, Q, when entering into a ferromagnetic material (F). This effect is described in the text. For simplicity a spherical Fermi surface is considerated. b) Oscillations of the induced superconducting order parameter in F as a function of the distance from the S/F interface, x_F. Negative values of the order parameter show up for particular ferromagnetic layer thicknesses.

The formalism of the superconducting proximity effect in the dirty limit [9], [11], [12] based on the Usadel equations [13], provides the microscopic background to fully account for the thermodynamics [14] and out-of-equilibrium properties of this inhomogeneous induced superconductivity. However the basic idea sketched above for a ballistic system remains valuable even when diffusion takes over as it may occur in actual samples [15].

To investigate this inhomogeneous superconducting state and its implications for Macroscopic Quantum Mechanics experiments hybrid F/S nanostructures are needed As the exchange energy of ferromagnetic d-elements is typically of 100 meV-1 eV, the penetration of Cooper pair in F is very short range ˜ 1nm. The controle of the interface quality at this length scale being still a challenge, a breakthrough in the field came when weak ferromagnetic alloys started to be used. Lower exchange energies insure larger ξ_F, compatible with standard deposition techniques. Two types of alloy have been successfully used: CuNi [16] and PdNi [17] with coherence lengths of 5-10 nm and Curie temperatures varying from few tens of Kelvin up to hundred Kelvin. So far, experiments with millimetric and sub micrometric sized tunnel and Josephson junctions have been performed. Tunnel junctions allow to measure the quasi-particle density of states (DOS) with unsurpassed energy and amplitude resolution while the Josephson effect addresses the current phase-relation ship between two weakly connected superconductors. Depending on their size, samples have been fabricated either by simple mechanical masking during evaporation or optical lithography or even electron lithography. Both e-gun evaporation and DC- magnetron sputtering have been used. The superconductor is usually Nb.

The oscillations of the superconducting wave function modify the coherent superposition of the quasi-particle states in F. As a consequence, when the superconducting wave function is negative the DOS is up-side-down [14] resulting in cap-sized tunneling spectra . This has been observed experimentally in Al/Al_2O_3/ PdNi/Nb junctions [17] the Al_2O_3 being an insulator layer working as a tunnel barrier. Of course the critical thickness to reach a negative order parameter depends on the Ni concentration. For about 10% Ni in Pd, this thickness is found to be 6 nm in good agreement with the estimation of the PdNi exchange energy (10 meV). The finite resistance at the PdNi/Nb interface on the order of $10^{-10}\ \Omega cm^2$ [18], reduces the leakage of Cooper pairs. For negative order parameters the weak coupling with a second superconductor reverse the sign of the Josephson current in the current-phase relationship $I=I_c \sin\varphi_{12}$, where I_c is the junction critical current and φ_{12} the phase difference between the two superconductors. Formally this is equivalent to a π-phase shift between the two superconductors, therefore this kind of junction is commonly called a π-junction. As the measurement of the critical current doesn't allow determining the sign of the current through the junction, the transition from 0-to-π junctions as a function of the ferromagnetic layer thickness is revealed as a zero in the junction critical current. Indeed, the critical current of in Nb/Al/Al_2O_3/ PdNi/Nb junctions measured from their current-voltage characteristics (I-V characteristics) follows the oscillations of the superconducting wave function induced in F and shown in fig.1b [19]. The ground state of a Nb/NbO/PdNi/Nb junction has been measured very recently by a Quantum Interference experiment using a mesoscopic superconducting loop [20]. Finally it must be stressed out that the first indication of π-coupling in S/F nanostructures, although controversial, was reported as the observation of an oscillating critical temperature in Gd/Nb multilayer [21].

A serie of experiments carried out in mesoscopic samples fabricated of Al and either Co [22] or Ni [23] has addressed the possibility of a long range proximity effect in a ferromagnetic finger connected to a superconducting reservoir. Even though the difficulties in preparing well controlled samples including a reliable geometry brought up serious doubts on the interpretation of those experiments, theoretical speculations have shown that a long range proximity effect may indeed exist if a triplet component of the induced superconducting order parameter is generated in F. As the exchange field is not pair-breaking for triplet superconductivity, the induced triplet component leak in F on length scale much larger than singlet Cooper pairs. Whether or not such a component can be generated either by an inhomogeneous magnetization at the interface, a domain wall or other mechanisms is currently under investigation [24]. However, as this state is very sensitive to spin-orbit scattering, its implementation requires ballistic ferromagnets. For this and other reasons, the realization of structures with a large spin mean free path represents a major challenge also in the future of the hybrid nano-structures.

Let conclude this part simply remarking that if the effect of the magnetic order on the superconducting wave function has made a step forward in the last decade, the effect of the superconducting codensate on the long range magnetic order remains an open question. Can hybrid nanostructures also help to answer to this question ?

Spin polarized transport in Superconductor/Ferromagnet junctions.

In the first part, I considered Cooper pairs entering into a ferromagnet and I neglected spin leakage from the ferromagnet into the superconductor. At the equilibrium, this is a quite good approximation as the superconducting reservoir is hardly polarized by the magnetic proximity effect on a length scale which is usually less than one atomic monolayer [25]. Therefore it may be completely hidden by interface roughness including mixing. However, the situation changes when a current is driven from the ferromagnet to the superconductor, as spins are forced to penetrate in S. Where they generate a spin imbalance and hence an out-of-equilibrium magnetization as shown in fig.2. Of course, the spin imbalance depends on the spin polarization of the ferromagnet and the current density through the S/F interface. Thus, contrarily to the superconducting proximity effect described above where weak Stoner ferromagnets have been used, spin transport studies have been carried out in materials with a high spin polarization. Semimetals provide a natural candidate. But how far from the interface goes this magnetization into the superconductor ?

We can answer to this question by steps. Let first see the spin drift if the superconductor were a normal metal (N). I will address the effect of superconductivity on it later. As spin current unlike charge current is not conserved, the spin current decays moving away from the F/N interface. Spin-orbit scattering and spin-flip scattering mix up the spin orientations and they reduce the spin mean free path. The time spent in N before undergoing to a non-conserving spin scattering is the spin relaxation time, τ_s. Of course, it doesn't mean that after τ_s the electron looses its spin. τ_s just defines for how long the spin remains a good quantum number in N. As spin-orbit scattering depends on the atomic number (Z) of the normal metal, the spin relaxation time is larger for light elements as Mg and Al [26]. If the transport in N is diffusive, i.e. the sample size is much bigger than the electron mean free path, the spin mean free path is given by $l_s = (D\tau_s)^{1/2}$ and is usually called spin diffusion length. For a Fermi liquid as a normal metal, D is the diffusion constant. Formally the diffusion of spins from F to N can be treated like Cooper pairs penetrating into a normal dirty metal, N . Note that, the exponential decay of the spin current as well as of the superconducting correlations in N results from diffusion. The spin relaxation time has been measured by the Hanle effect [27]. The out-of equilibrium magnetization can be probed connecting the normal metal with other ferromagnetic electrode. Depending on the magnetization orientation of this electrode, a voltage proportional to the out-of equilibrium magnetization appears at the second N/F interface. Measurements at low temperature on an Al single crystal using a SQUID pico-meter have found τ_s ~ 1ns and a very large spin diffusion length l_s ~ 500 μm [28], [29].The same experiments on Au [30] thin films lead to a much shorter τ_s ~ 10 ps as expected and in agreement with weak localization data. The spin diffusion length depends on the mean free path and was found l_s ~ 1.5 μm in those films. Recently, the spin diffusion length in Cu [31] and Al [32] mesoscopic samples has been directly obtained by probing the out-of-equilibrium magnetization at different distances from the F/N interface on the same sample. The debate on the discrepancies with previous results is still open.

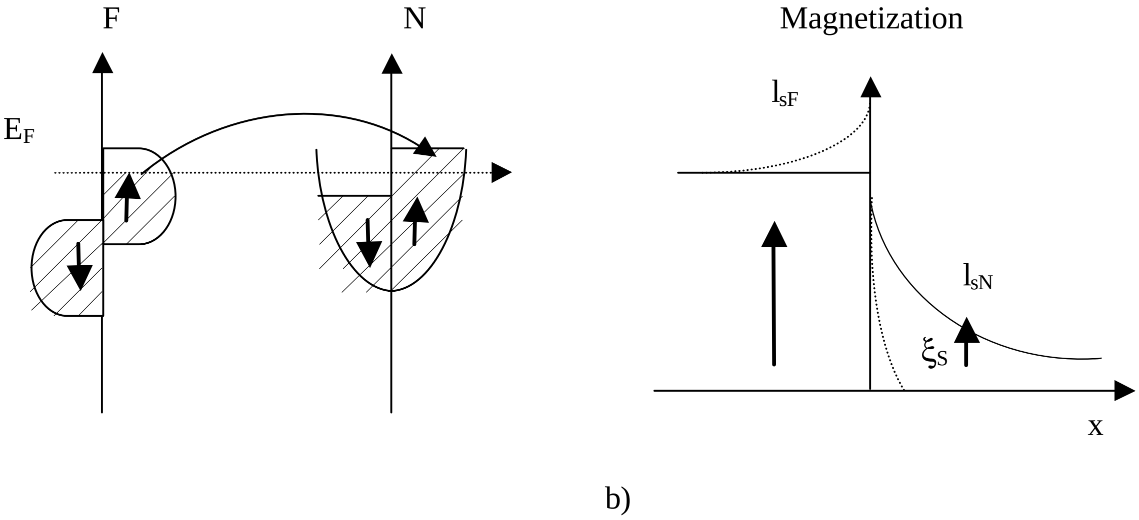

Fig.2 : a) Diagram of the density of states of a ferromagnetic/normal metal (F/N) junction.. The spin imbalance is generated in N by spin injection. Here the ferromagnet is 100 % spin polarized. b) The out-of-equilibrium magnetization resulting from the spin imbalance is illustrated at the F/N (solid line) and F/S (dotted line) boundaries. l_{sN} and l_{sF} are the decay lengths of the spin accumulation regions in N and F respectively. x_S is the superconducting coherence length.

In a superconductor the situation is in principle different. In fact, no spin current can be carried by the condensate as Cooper pairs have zero spin. Therefore spins can penetrate only if their energy is larger than the paring energy, Δ_s. This requires a potential drop at the F/S interface larger than Δ_s/e, where e is the electron charge, otherwise the spin is scattered back in F. thus, spins penetrate in S only over ξ_F, the superconducting coherence length. The number of backscattered electrons depends on their polarization. For a 100 % polarized ferromagnet all the electrons must be scattered back as no minority spins are available to form Cooper pairs. They accumulate in F at the F/S interface on a length scale given by the spin diffusion length in F [33] (see fig.2b). Thus the resistance of a F/S diode at low bias increases with increasing polarization. This property has been used to probe the spin polarization by point contact (also called Andreev) spectroscopy. Two types of F/S diodes have been realized. One consisting of a small superconducting tip (Nb) in contact with F [34], the other one based on a S/F nanocontact obtained by evaporation through a nano-holed Si_3N_4 membrane [35]. The measured polarization rates (0.4 for Co, 0.32 for Ni …) are consistent with previous tunneling experiments [26]. The quality of the contact as well as local spin excitations [36] may introduce aging factors. At larger bias, spin polarized electrons can be injected in S as “hot electrons”. Quasiparticles can carry a spin current. This spin polarized quasiparticle imbalance in S relax into the condensate. Whether or not during the relaxation the spin accumulation affect the superconducting state depends on the characteristic relaxation times for spin and charge. A much larger charge relaxation time would basically lead to a greater number of out-of-equilibrium quasiparticles than spins, making negligible the effect of the magnetization on the condensate. Only few theoretical works have address this topic that remains experimentally unexplored.

The situation may be different in unconventional superconductors. The pairing energy going to zero at some point of the Fermi surface, spins can enter in S even at zero energy. However, how these spins couple to the condensate is not very well understood. Recent experiments on high Tc superconductors have shown that the critical current of a HTCs bridge can be controlled by spin injection [37]. The device was a YBCO/LSCO bilayer, where the maganite is supposed 100% spin polarized. Although it is not completely clear whether or not unwanted heating effects can be definitely excluded [38], theoretically it has been pointed out that an out-of-equilibrium magnetization affects more severely unconventional than conventional s-wave superconductors [39]. Thus, spin injection may be used as current bias in S/F switches. Although some theoretical works have stressed out as S/F based devices may provide high efficiency spin valves [40] [41], the field of out-of-equilibrium spin imbalance in superconductors remains quite unexplored.

Finally an even more fundamental question rises. Can spin transport measure only spins and not charges? If so, spin injection would provide a powerful technique to investigate charge-spin separation in strongly correlated electron systems.

Acknowledgments. I thank B. Leridon, W. Guichard, T. Kontos, J. Lesueur, M. Moragues and ML. Della Rocca for many fruitful discussions.

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