Proximity effects in the superconductor / heavy fermion bilayer system Nb / CeCu₆.

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Abstract. – We have investigated the proximity effect between a superconductor (Nb) and a 'Heavy Fermion' system (CeCu₆) by measuring critical temperatures $T_c$ and parallel critical fields $H_{c2}(T)$ of Nb films with varying thickness deposited on 75 nm thick films of CeCu₆, and comparing the results with the behavior of similar films deposited on the normal metal Cu. For Nb on CeCu₆ we find a strong decrease of $T_c$ with decreasing Nb thickness and a finite critical thickness of the order of 10 nm. Also, dimensional crossovers in $H_{c2}(T)$ are completely absent, in strong contrast with Nb/Cu. Analysis of the data by a proximity effect model based on the Takahashi-Tachiki theory shows that the data can be explained by taking into account both the high effective mass (or low electronic diffusion constant), and the large density of states at the Fermi energy which characterize the Heavy Fermion metal.

Introduction. – When a superconductor (S) is brought into contact with a non-superconducting conductor (N), superconductivity leaks into that material by the proximity effect [1]. For the thermodynamic properties of the S/N bilayer such as the critical temperature $T_c$ or the upper critical field $H_{c2}$, this can be modelled as a spatial variation of the superconducting pair density. The spatial variation mainly depends on the electron diffusion constants for the S- and the N-metal, on the transparency of the interface for electrons and Cooper pairs, and on the pair breaking mechanisms on the N-side of the interface. Until now, two classes of N-materials have been investigated. The first is formed by simple metals such as Cu or Au, where the physics is mostly understood. Pairs are broken at finite temperatures by thermal fluctuations, leading to dephasing of the constituents of the induced Cooper pair. This is translated into
a temperature-dependent characteristic length over which superconducting correlations penetrate, the ‘normal metal coherence length’ $\xi_N$. Since $\xi_N = \sqrt{\hbar D_N/(2\pi k_B T)}$ ($D_N$ is the diffusion constant of the N-metal, $T$ is the temperature, the other symbols have their usual meaning), this length can become large at low temperatures. In the absence of other pair breakers, $T_c$ of a bilayer with finite N-layer thickness $d_N$ will be finite. The other class of materials is formed by ferromagnets (F) such as Fe; pair breaking is due to the exchange interaction $E_{ex}$ which acts on the spins of the Cooper pair. For strong magnets ($E_{ex} >> k_B T_c$) it results in a temperature-independent $\xi_F = \sqrt{\hbar D_F/(2\pi E_{ex})}$ with a characteristic value of only a few nanometers. In this case, superconductivity can already be quenched at a finite value for $d_S$, called the critical thickness $d_S^c$.

Little attention has yet been paid to N-layers where the electronic ground state is dominated by many body correlations, such as in a Heavy Fermion (HF) metal. Basically, HF metals consist of a lattice of atoms with localised (f-)electrons, where the magnetism is quenched by a coherent Kondo effect. This leads to a strong peak in the DOS near the Fermi energy with small energy width (for relevant reviews, see [4, 5]). The consequences for the low temperature physics can be phenomenologically described in terms of Landau Fermi liquid theory by a mass renormalization of the charge carriers. The ensuing large effective mass $m^*$ can be directly observed in e.g. the specific heat of the system, $c_v$, where the linear term $\gamma = c_v/T \propto m^*/m_e$ (with $m_e$ the bare electronic mass) can be up to two orders of magnitude larger than in normal metals. Since the magnetic moments are often not completely quenched, HF systems can also show magnetic order at low temperatures. For the proximity effect in an S/HF system, several scenarios are possible. In the spirit of the Fermi liquid approach, the HF metal can be considered a normal metal with a large mass, or equivalently a low Fermi velocity and therefore a small diffusion constant. Also, additional pair breaking mechanisms may be present due to the strong electron-electron interactions or the residual moments. On the other hand, the interface transparency may be decreased due to the mismatch in Fermi velocities, which would shield the superconductor from the HF metal and counter the other two effects.

In order to investigate these questions, we have performed a comparative proximity effect study of the thin film systems Nb/CeCu$_6$ and Nb/Cu. CeCu$_6$ is a well-known HF system, with an extremely large value of $\gamma \approx 1.6$ J/(mole K$^2$) [6] and no magnetic order down to the mK-regime, making it a good model system for this study. Nb/Cu is a much studied proximity system which shows one feature of particular interest, namely a Dimensional Crossover (DCO) from three-dimensional (3D) to two-dimensional (2D) behavior in the temperature dependence of the critical field parallel to the layers $H_{c2}^{\parallel}$ [7, 8]. The DCO occurs when the N-layer thickness becomes of the order of $\xi_N$, and can be modelled quite accurately. In particular, recent calculations by Ciuhu and Lodder based on the Takahashi-Tachiki theory for metallic multilayers [9], and including a finite interface resistance $R_B$, showed that quantitative agreement between theory and experiment can be found in the case of Nb/Cu for very reasonable values of the different parameters [10]. Here we present data on $T_c(d_S)$ and $H_{c2}^{\parallel}(T)$ for bilayers Nb/Cu and Nb/CeCu$_6$ and compare them with similar calculations. We show that the HF system can be treated as a normal metal, with both the low diffusion constant (Fermi velocity) and the high DOS as necessary ingredients to explain the observed behavior.

**Experimental.** – Sets of CeCu$_6$ films and bilayers of sub/CeCu$_6$/Nb (sub denotes the substrate) were fabricated by DC-magnetron sputtering in an ultra high vacuum (UHV) system with a background pressure of the order of $5 \times 10^{-10}$ mbar, and an Ar sputtering pressure of $2.5 \times 10^{-3}$ mbar. Crystalline CeCu$_6$ was grown as reported before [11], at a temperature of 350 °C, using Si-substrates with amorphous Si$_3$N$_4$ buffer layers to prevent Cu diffusion at those
Fig. 1 – Materials characteristics of the CeCu$_6$ films. (a) RBS spectrum (nr. of counts versus backscatter energy $E_{BS}$) taken with $^4$He-ions of 2 MeV on a sample Si/Si$_3$N$_4$/(75 nm CeCu$_6$)/(15 nm Nb). The different elements are indicated. The thin smooth line is a fit to the measured curve. (b) Specific resistance as function of temperature for single CeCu$_6$ films of 75 nm (o) and 100 nm (△).

temperatures. The Nb was deposited on top of the CeCu$_6$ after cooling the substrate holder with cold nitrogen gas to close to room temperature. Composition, thickness and crystallinity of the films were determined by Rutherford BackScattering (RBS) measurements together with X-ray diffraction measurements at low and high angles. The RBS measurements show good agreement with the expected stoichiometry for CeCu$_6$ and no diffusion is found either of Ce or Cu into the substrate or of Nb into the CeCu$_6$. Fig. 1a shows part of the RBS spectrum for CeCu$_6$(75 nm) / Nb(15 nm) on a Si/Si$_3$N$_4$ substrate and a fit of the data without taking any diffusion into account. Bilayers and trilayers of sub/Cu/Nb and Cu/Nb/Cu were grown in a different UHV system with similar background pressure and sputtering conditions. In order to compare results, $d_{Nb}$ in the trilayers was taken two times $d_{Nb}$ in the bilayers, which yields equal conditions for the superconducting order parameter in the middle of the film (for the trilayer) and at the vacuum interface (for the bilayer). Resistance measurements were performed in standard 4-point geometry on lithographically patterned samples with bridge widths of 200 $\mu$m and a distance of 1.2 mm between the voltage contacts. The electrical resistivity $\rho$$_{CeCu6}$ as function of temperature for two single films is shown in Fig. 1b, and behaves as reported before [11], with a clear maximum in $R$(T) at $T_{max} \approx 5$ K, similar to what is found for bulk material [12], and a residual resistivity of the order of 80 $\mu$Ωcm. We decided to use 75 nm thick CeCu$_6$ layers, which with $T_{max}=4$ K suggest only little deviation from the bulk properties.

Results and discussion. – The dependence of $T_c$ on $d_{Nb}$ for the bilayer set sub/CeCu$_6$(75 nm)/Nb($d_{Nb}$) is shown in Fig. 2a, and compared to that of single Nb films, prepared in both UHV systems. For the Nb films a slight suppression is witnessed, usually found in the case of Nb, and caused by a mixture of different effects such as oxidation through the unprotected top layer, a small proximity effect with the substrate, and lifetime broadening due to growth disorder [14]. For the S/HF bilayers, the suppression is much stronger, with a critical thickness $d_{cr}$ for onset of superconductivity reached around 12 nm. This would be equivalent to 24 nm in a trilayer configuration, and is of a similar magnitude as found in superconductor/ferromagnet systems such as V/Fe and Nb/Fe [15,16]. The result shows immediately that the interface does allow particle exchange, so the supposed huge Fermi velocity mismatch in the system of order...
Fig. 2 – (a) $T_c$ as function of Nb thickness $d_{Nb}$ for bilayers $sub/CeCu_6/Nb$ (●) and single Nb films (+). The drawn line is a fit using the proximity effect model. The dotted line is a model calculation with $N_{CeCu_6} = N_{Cu}$. The inset shows the behavior of $D_{Nb}$ as found from the fit. (b) Parallel critical field $H^\|_{c2}$ as function of $T$ for a bilayer $sub/Cu/Nb$ ($\bigtriangleup$; $d_{Nb} = 15$ nm) and a trilayer $Cu/Nb/Cu$ (○; $d_{Nb} = 30$ nm). Drawn lines are model fits. (c) $H^\|_{c2}$ as function of $T$ for bilayers $sub/CeCu_6/Nb$ with thicknesses $d_{Nb} = 13.5$ nm (lowest $T_c$), 15 nm, 17 nm, 25 nm, 30 nm, 50 nm. Drawn lines are model fits. (d) Model calculation of the inverse slope of $H^\|_{c2}$ at $T_c$ versus the effective electron mass $m^*$. $10^4$ does not lead to a highly reflective interface. It also shows that the coherence length in the HF-material $\xi_{HF}$ must be small, leading to the strong suppression.

Another indication of behavior deviating from simple S/N systems comes from the parallel critical field $H^\|_{c2}(T)$. To demonstrate the difference, Fig. 2b shows $H^\|_{c2}(T)$ for a bilayer of $sub/Cu(75 \text{ nm})/Nb(15 \text{ nm})$ and a trilayer of $Cu(75 \text{ nm})/Nb(30 \text{ nm})/Cu(75 \text{ nm})$, to be compared to the data of the S/HF bilayers given in Fig. 2c. Decreasing $T$ near $T_c$, $H^\|_{c2}(T)$ is linear for the Nb/Cu samples, followed by a kink and $\sqrt{1-T/T_c}$-like 2D behavior. The kink signals the well-known DCO [7,8], usually observed in multilayers, but also present in tri- and bilayers. In strong contrast, the S/HF bilayers do not show a DCO but only 2D-behavior for all $d_{Nb}$. Qualitatively, this again indicates a small value for $\xi_{HF}$: the superconducting order parameter does not penetrate sufficiently far into the HF metal to yield a coupled system.

To make more quantitative statements, we analyzed the data by means of model calculations based on the Takahashi-Tachiki formalism. Details are given in ref. [10], but we reiterate the main elements in order to introduce the different parameters. The formalism solves the equation for the pair potential $\Delta(\mathbf{r})$ with a space-dependent coupling constant $V(\mathbf{r})$ for small
\[ \Delta(r) = V(r)k_B T \sum_{\omega} \int d^3r' Q_\omega(r, r') \Delta(r') \]  (1)

in which \( Q_\omega \) is an integration kernel still to be determined, \( V(r) \) is the BCS coupling constant and the other symbols have their usual meaning; the summation runs over the Matsubara frequencies. In the dirty limit, \( Q_\omega \) can be shown to obey:

\[ [2|\omega| - \hbar D(r)(\nabla - \frac{2ie}{\hbar c}A(r))^2]Q_\omega(r, r') = 2\pi N(r)\delta(r - r') \]  (2)

with \( N(r) \) the DOS at the Fermi energy and \( D(r) \) the diffusion constant. These equations are complemented with boundary conditions for \( \Delta \) at the interface which parametrize a possible barrier encountered by the electrons through a boundary resistivity \( R_B \). In the calculations we use fixed values for the density of states ratios \( N_{Cu}/N_{Nb} = 0.16 \) and \( N_{CeCu6}/N_{Nb} = 320 \). The former value was also used in ref. [10] and derives from ref. [15]; the latter value is constructed from the former by the ratio \( \gamma_{CeCu6}/\gamma_{Cu} = 2000 \). The value of \( T_{c,Nb} \) was fixed at 9.2 K, except for the Nb/Cu bilayer with \( d_{Nb} = 15 \text{ nm} \) where it was 8.2 K. This reflects the fact that \( T_c \) for thin Nb-layers starts to decrease, as explained above. Fitted were the different diffusion constants \( D_N \) and boundary resistances \( R_B \). The results of the fits for the critical field data are shown in Fig. 2 as solid lines. The parameter values are given in Table II. For the two Nb/Cu samples, the fitted values are in very reasonable agreement with the numbers found by fitting the data of Chun et al. [8,10]. The values for \( D_{Nb} \) are roughly equal, the values for \( D_{Cu} \) are slightly higher, which probably reflects the difference in preparation conditions, and also the values for \( R_B \) are very similar [10]. For the Nb/CeCu6 samples it can be noted that \( D_{Nb} \) increases slowly and more or less linearly with increasing \( d_{Nb} \); using this linear variation, which is plotted in the inset of Fig. 2, and values of \( D_{CeCu6} = 0.1 \times 10^{-4} \text{ m}^2/\text{s} \), \( R_B = 324 \times 10^{-8} \mu \Omega \text{cm}^2 \) we calculated the behavior of \( T_c(d_s) \) as a consistency check. The agreement, shown in Fig. 2a, is equally satisfactory. The most interesting values from the fits are of course those for \( D_{CeCu6} \), which are much lower than those for Cu. Assuming a Fermi velocity of the order of \( 10^3 \text{ m/s} \), a value for \( D_{CeCu6} \) of \( 0.1 \times 10^{-4} \text{ m}^2/\text{s} \) yields a mean free path \( l_c \) of 3 nm, which is not surprising in view of the strongly granular nature of the films. Still, a low value for \( D_{N} \) by itself does not necessarily reflect the heavy Fermion character: if \( CeCu6 \) is taken as a Cu matrix strongly diluted by a small amount of Ce atoms, with a mean free path of the order of the interatomic distance, \( D \) would also be very small, of order \( 10^{-4} \text{ m/s} \). However, as we show now, just a low value for \( D_{N} \) cannot describe the measurements. To
demonstrate this, we calculated $T_c(d_S)$ for Nb/CeCu$_6$ with the parameters given above, but with the DOS-ratio for Nb/Cu, thereby mimicking a very dirty but otherwise standard N-metal. The result, shown in Fig. 2, as dotted line, shows that $T_c$ now drops at much lower $d_{Nb}$. This can be understood by realizing that the low diffusion constant inhibits penetration of Cooper pairs in the N-metal and therefore also inhibits pair breaking, leading to a smaller amount of suppression of $T_c$. What makes the difference is the high DOS-value for CeCu$_6$: the large number of available states works as a sink for Cooper pairs which counteracts the low diffusion constant. Our major conclusion is therefore that both the low $D_N$ and the high $N_N$ are necessary ingredients in the description of the data. This leads to the question what the effective mass needs to be in order to suppress the DCO which is so characteristic for an S/N system. For this we calculated values for $dT/dH^\parallel|_{T_c}$, the inverse parallel critical field slope at $T_c$, as function of the effective mass $m^*/m_e$ ($m_e$ being the bare electron mass) as used in the free-electron expressions for $N_N = \frac{m^* k_F}{\hbar^2}$ and $D_N = \frac{k_F l_e^3}{m^*}$, with $k_F$ the Fermi wave vector. As shown in Fig. 2, low values for $m^*$ yield a finite value for the inverse slope, signifying 3D behavior and therefore a DCO, which goes to 0 around $m^* = 10$, meaning that the DCO has disappeared and 2D behavior has set in. Clearly, CeCu$_6$ is far into this regime.

Finally, we come back to the difference in proximity effects between F-, and HF-metals. Using the expression for $\xi_N$ given in the Introduction at a typical value of $T = 5$ K and with the fitted values for $D_{CeCu_6}$, we find $\xi_{CeCu_6} \approx 1.5$ nm. This is very similar to values found for strong ferromagnets, but the physics of the strong suppression of superconductivity which is found both in the F- and HF-systems is different. In the F-case, the small value of $\xi_F$ derives from the large pair breaker ($E_{ex}$), in the HF-case from the small $D_N$. As shown in Fig. 2, a low value for $D_N$ does not yield strong suppression of superconductivity, that is actually due to the high value for $N_N$. This emphasizes once more that the basic proximity-effect parameter is not $\xi_{F,HF}$ but rather $\gamma = (\rho_S \xi_S)/(\rho_X \xi_X)$ with $X = F$, HF [3, 17]. Since $\rho_X \propto (N_X D_X)^{-1}$ and $\xi_X \propto \sqrt{D_X}$ it can easily be seen that for the F-system $\gamma$ is large due to $E_{ex}$, and for the HF-system $\gamma$ is large due to $N_{CeCu_6}$. For the HF-system, an extra pair breaker is not needed in the description.

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