Eur. Phys. J. B 1, 405–407 (1998)

THE EUROPEAN
PHYSICAL JOURNAL B

Proximity effect in Fe/Pb/Fe trilayers

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Received: 30 June 1997 / Revised: 20 August 1997 / Accepted: 16 December 1997

Abstract. We have studied superconducting and magnetic properties of sputtered Fe/Pb/Fe-trilayers. For a fixed Pb thickness d_{Pb} and with changing Fe thickness, d_{Fe}, a monotonic decrease of the superconducting transition temperature T_{c} was observed. Magnetization measurements clearly showed that Fe remains ferromagnetic down to the monolayer range. A quantitative comparison of $T_{\mathrm{c}}(d_{\mathrm{Fe}})$ with the theory of pair breaking by the exchange field reveals that the observed T_{c}-suppression by the ferromagnetic Fe-layer is much weaker than expected. Possible reasons for the reduced T_{c}-suppression in this system are discussed.

PACS. 74.80.Dm Superconducting layer structures: superlattices, heterojunctions, and multilayers – 74.50.+r Proximity effects, weak links, tunneling phenomena, and Josephson effects – 74.62.-c Transition temperature variations

Motivated by the significant progress achieved in the preparation and study of metallic multilayer system in recent years [1], a renewed interest in proximity effects of superconductor/ferromagnet (S/F) multilayers is noticeable (see *e.g.* [2] and references therein). Although it is a classic topic, until now the proximity effect at an S/F interface is far from being quantitatively understood. The main experimental problem is the real interface structure in metallic multilayer system which often does not exhibit sharp S/F transitions and uniform properties on either side of the interface. In this context it is important to study S/F multilayer systems with sharp interfaces without interdiffusion and with properties of the S and F single layers close to the properties of the bulk material.

Here we report preliminary results on the study of a Fe/Pb/Fe trilayer system. We have found that Fe/Pb layered system can be considered as a first example of an S/F system with 'ideal' interfaces. Actually this system was among the first in which the S/F proximity effect has been studied. Hauser *et al.* [4] have observed a decrease of T_{c} with decreasing lead thickness d_{Pb} at two fixed iron thicknesses d_{Fe}. The investigation of $T_{\mathrm{c}}(d_{\mathrm{Fe}})$ for a series of Fe/Pb/Fe trilayer samples is the main purpose of our present work.

The samples were prepared by rf sputtering techniques on $\mathrm{Al_2O_3}$ $(11\bar{2}0)$ substrates at room temperature. Pure Ar (99.999%) at a pressure of 5×10^{-3} mbar was used as a sputter gas. Pure Pb (resistivity ratio RRR = R(300 K)/R(10 K) = 50) and Fe (99.99%) targets were used for deposition.

For a single Pb film with a thickness of 600 Å, as used for our present trilayer systems, we observe the bulk value for superconducting (SC) transition temperature $T_{\mathrm{c}} \simeq 7.2$ K and a residual resistivity of 2 $\mu\Omega$ cm.

In order to study the dependence of the SC parameters on the thickness d_{Fe}, we prepared a series of 9 different Fe/Pb/Fe trilayer samples with different d_{Fe} at a constant value of d_{Pb} within one single run, using a shutter system. The nominal thicknesses of the Fe-layers in the trilayer series varied between 4 Å and 30 Å, as determined by the deposition rate and verified by quantitative wave length dispersive electron microprobe analysis.

Using a SQUID magnetometer we measured magnetic hysteresis loops at $T = 20$ K with the film surface parallel to the direction of the magnetic field. The hysteresis loops exhibit a square shape, typical for ferromagnetic (FM) Fe films without any noticeable change with varying Fe-thickness. We have found that the saturation magnetization is independent of d_{Fe} down to at least $d_{\mathrm{Fe}} = 6$ Å. For the samples with $d_{\mathrm{Fe}} = 4$ Å a FM hysteresis loop could clearly be resolved but an absolute value for the saturation magnetization M cannot be evaluated due to a large uncertainty in the determination of d_{Fe}. This clearly indicates that no nonmagnetic or non ferromagnetic Fe-layer exists in the Pb/Fe interface. In this sense Pb/Fe system has a "sharp" interface, in contrast to the case of

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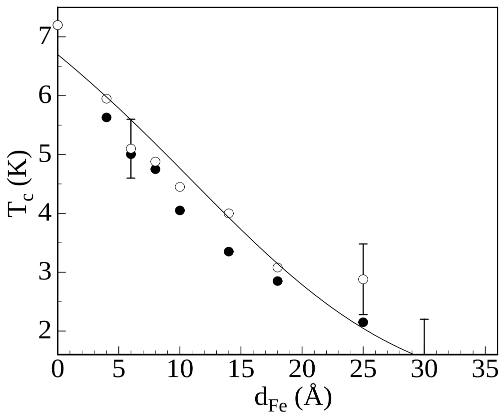

Fig. 1. T_c *versus* d_{Fe} as determined by ac susceptibility (closed symbols) and resistivity (open symbols) for samples with fixed $d_{Pb} = 620$ Å. The solid line is the fit using equations (1, 2, 4) with parameters given in the text.

Nb/Fe where a broad intermixed region at the interface was found previously [2].

The ferromagnetism of the Fe-layer was studied in addition by ferromagnetic resonance measurements at 9.4 GHz and at room temperature. We detected a sharp resonance signal down to about $d_{Fe} \simeq 10$ Å, for lower d_{Fe}-values we observed a strong broadening of the resonance line. Increasing of the FMR linewidth and disappearance of the resonance signal at $d_{Fe} = 4$ Å may be caused by the dispersion of the effective magnetization due to an initial island growth of the iron layer and due to a long scale roughness of the Fe layers.

We have measured T_c resistively and by means of ac magnetic susceptibility measurements. In both cases the temperature corresponding to half the value of the maximum transition signal was defined as T_c. The observed sharp transitions confirm high quality of our films. The resulting $T_c(d_{Fe})$-dependence is shown in Figure 1. A monotonic depression of T_c is observed up to d_{Fe} =25 Å, for d_{Fe} = 30 Å T_c is below the experimental temperature range.

Bearing in mind our magnetization data we conclude that the T_c-depression in the Pb/Fe system occurs due to the penetration of Cooper pairs through the interface into the Fe-side, where they are subjected to a pair-breaking effect of the magnetic exchange field. Quantitatively the efficiency of the pair breaking is determined by the exchange field I and by the interface "transparency" parameter, $\eta < 1$ describing the relative density of the Cooper pairs in the Fe-layer. We performed calculations of $T_c(d_{Fe})$-dependence taking into account the finite penetration length of the pair density into the FM layer using the standard equation for $t_c = T_c/T_{c0}$ (here T_{c0} is the transition temperature for a single SC layer)

$$\mathrm{Re}\,\Psi\left(\frac{1}{2}+\rho\right) - \Psi\left(\frac{1}{2}\right) + \ln(t_c) = 0, \tag{1}$$

where $\Psi(x)$ is the digamma function. Following Radović *et al.* [3], who have calculated the T_c-suppression in S/F multilayers, the pair breaking parameter with slight modifications can be expressed as

$$\rho = \frac{\eta k_M \xi_S^2}{d_S}\frac{1}{t_c}\tanh(k_M d_M), \tag{2}$$

where
$$k_M = \left(\frac{2iI}{\hbar D_M}\right)^{1/2}. \tag{3}$$

Here d_S and d_M are the thicknesses of SC and FM layers, respectively, ξ_S is the coherence length in the SC, and D_M is the diffusion coefficient in the FM layer. Consistent with Radović *et al.* [3], one can find that equations (1–3) predict highly nonmonotonic dependencies of T_c on d_M, with a deep mimima around $d_M \sim |k_M|^{-1}$, which is in sharp contrast to our experimental data (see Fig. 1). In fact, equations (2, 3) have to be modified taking into account spin-orbit scattering, which is known to be strong in Pb. Very roughly the latter can be included phenomenologically by replacing the k_M-value in the pair-breaking parameter by

$$k_M = \left(\frac{2iI}{\hbar D_M}\frac{1}{1+i\hbar(I\tau_{so})^{-1}}\right)^{1/2}, \tag{4}$$

where τ_{so} is the spin-orbit scattering time [5]. Solving equation (1) with the new k_M, we found that the peculiar nonmonotonic behavior of the $T_c(d_M)$-dependence completely disappears in the limit of a low spin-orbit scattering time, and the similarity between calculated and experimentally observed $T_c(d_M)$-dependence becomes more obvious [6].

In order to fit the observed T_c-dependence by equations (1, 2, 4), we first estimate the SC coherence length ξ_S. Using the value of the mean free path $l \simeq 200$ Å for the Pb layer, derived from our residual resistivity measurements, and $\xi_{BCS} \simeq 830$ Å, the SC coherence length ξ_S can be obtained as [3] $\xi_S = \sqrt{(\xi_{BCS} l)/3.4} \simeq 200$ Å.

We find that with any reasonable choice of the ratio $(I/\hbar D_M)$ one needs a very low transparency parameter η to fit our $T_c(d_M)$ data. The best fit of the theoretical curve (solid line in Fig. 1) yields the 'coherence length' in the FM layer $(\hbar D_M/I)^{1/2}$ =23 Å, and $I\tau_{so} = \hbar$, $\eta \simeq 4\times10^{-2}$. Assuming $I \sim 1$ eV in Fe, one then estimates $\tau_{so} \sim 10^{-15}$ s, which appears to be unusually low. However, one should keep in mind that the scattering takes place at the interface where spin-orbit scattering must be very strong due to the large difference in the atomic numbers for Pb and Fe. In addition, static fluctuations of the exchange splitting near the interface are expected to be large, giving an additional contribution to the spin scattering.

We need a very low transparency η for the explanation of the weak T_c-suppression in the experiment explained. This reduction of the proximity effect might be due to a thin oxide interlayer at the interface. However from an experimental point of view we would exclude this possibility. We believe that the primary reason is a weak hybridization of the Pb and Fe derived wave functions at the interface, as it can be suggested by the absence of any solubility in the Fe-Pb phase diagram. This will strongly reduce the pair density in the Fe-layer. The second reason for the low transparency parameter is a strong reflection of the conduction electrons at the potential step of the interface, which will further reduce the Cooper pair density in the Fe-layer.

We would like to thank M. Brinkmann for his help in the computer calculations. The support of this work by the Deutsche Forschungsgemeinschaft (DFG-ZA161/6-2) and by the Russian Fund for Fundamental Research (Project 96-02-16332a), is gratefully acknowledged. One of us (I.A.G.) acknowledges NATO support under the Collaborative Research Grant HTECH.CRG961371.

References

1. E.G. Bauer *et al.*, J. Mat. Res. **5**, 852 (1990).
2. Th. Mühge *et al.*, Phys. Rev. B **55**, 8945 (1997).
3. Z. Radović *et al.*, Phys. Rev. B **44**, 759 (1991), and references therein.
4. J.J. Hauser, H.C. Theurer, N.R. Werthamer, Phys. Rev **142**, 118 (1964).
5. With k_{M} given by equation (4) the pair breaking parameter ρ (Eq. (2)) at $k_{\mathrm{M}} d_{\mathrm{M}} < 1$ interpolates between two well known limits: $\rho \sim iI$ in a weak spin-orbit coupling case, $(\hbar^{-1} I \tau_{\mathrm{so}}) >> 1$, and $\rho \sim I^2 \tau_{\mathrm{so}}/\hbar$ at the opposite limit.
6. It is worth to note that the oscillations of T_{c} due to to nontrivial phase difference $0 < \phi \leq \pi$ between neighbouring SC layers in F/S multilayers predicted by Radović *et al.* [3] seems to be also suppressed by the spin-orbit scattering.