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The electron mean free path and superconductivity in Cu/Pb modulated films

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Abstract. The electron mean free path L in the normal state and the critical temperature T_c as well as the parallel critical magnetic field $H_{c\parallel}$ and perpendicular critical magnetic field $H_{c\perp}$ were measured in equilayered Cu/Pb modulated films. The modulation period λ varied from 59 to 1620 Å. Our films were fabricated by thermal evaporation. The electron mean free path in the component layers estimated following the Gurvitch model is markedly shortened as a result of electron scattering both at interfaces and at grain boundaries. The dependence of T_c on component layer thickness was compared with existing theoretical models. Our measurements of the parallel critical fields as a function of temperature revealed the crossover of superconductivity from three-dimensional to two-dimensional behaviour.

1. Introduction

Artificial modulated films have been of great interest for almost 20 years. The possibility of a wide selection of component materials such as normal metals, ferromagnets, insulators, semiconductors and superconductors enables one to study carefully various phenomena in modulated structures. A detailed review has been given in [1–4].

From among many systems the layered artificial structures in which at least one of the constituents is a superconductor are still being analysed intensively. Their properties are especially interesting, because the thickness of component layers may be comparable with various length scales which characterize the superconducting state. One can distinguish three categories of superconducting multilayers depending on the separating material:

- (1) insulator (semiconductor) [5–10];
- (2) normal metal (superconductor with lower critical temperature) [11–17];
- (3) magnetic metal [18–23].

The main reason for the interest in these structures is the possibility of achieving a higher transition temperature into the superconducting state and an enhanced parallel critical field. Also the proximity effect used to probe for superconductivity in metals which have not displayed superconductivity as well as the existence of two antagonistic orders of ferromagnetism and superconductivity were studied in modulated superconducting films.

In the past we have investigated Ni/Pb films. We analysed their magnetic [24–26] and transport [27] properties and pointed out that symmetrical ($d_{\text{Ni}} = d_{\text{Pb}}$) multilayers did not display a superconducting state down to 1.3 K. To eliminate the influence of the ferromagnetism of the Ni layers on the Pb superconducting layers, we decided to prepare a series of Cu/Pb modulated films and to investigate their superconducting properties. The

Cu/Pb system seems to be a very good candidate for such investigations. A favourable feature of the Cu/Pb films is the very small interdiffusion and the lack of intermetallic compounds [28].

In this paper we report the superconducting properties of Cu/Pb modulated films. The outline is as follows. In the next section the details of the deposition procedure and measurement techniques are described. The transport properties of Cu/Pb modulated films and the relations of the critical magnetic fields to the temperature and of the critical temperature to the modulation period are discussed in section 3.

2. Experiment

The Cu/Pb equi-layered films (the component layers have equal thicknesses: $d_{\text{Cu}} = d_{\text{Pb}}$) were grown by alternating thermal evaporation of Cu and Pb from two independent crucibles. The glass substrates were water cooled during the evaporation procedures. The base pressure was 3×10^{-7} Torr and increased to 7×10^{-7} Torr during growth of the film. The deposition rates and the thicknesses of the component layers were measured by the use of quartz monitors. These rates for Cu and Pb were kept in the range of 5 \AA s^{-1} . Moreover, the total thickness of the modulated films was controlled after evaporation. It ranged from 4000 to 7000 \AA . To avoid the surface superconductivity the outer layers were always Cu. The shape of a film appropriate for transport measurements was obtained by the use of a mask.

The standard DC four-probe technique was used to measure the critical temperature as well as the parallel and perpendicular critical fields. The values were taken as critical when the extrapolated resistance of the samples reached zero. The measurements were performed in the gas environment in a specially designed set-up, immersed in a conventional ^4He cryostat, at temperatures down to 1.3 K.

The modulated structure of the Cu/Pb films was confirmed by the peaks of small angle x-ray scattering. A typical diffraction pattern for the sample with the modulation period $\lambda = 123 \text{ \AA}$ is shown in figure 1. The lattice mismatch, about 30%, is too large to allow for the observation of large-angle superlattice peaks [29]. The modulation period was also evaluated by dividing the total thickness of the samples by the number of bilayers. Both these methods for λ estimation were in good mutual agreement.

3. Results and discussion

3.1. Resistivity and electron mean free path

In this section the results of the resistivity measurements are presented. The evaluation of the resistivities of the component layers and the electron mean free path in modulated films following the Gurvitch [30] model is highly essential and useful in the interpretations of various measurements performed on metallic multilayers. These values are used for comparison of experimental results with the theoretical dependence of the critical temperature (see section 3.3). Below a temperature of 5 K, all the investigated Cu/Pb modulated films become superconducting. The changes in the critical temperature and critical fields with modulation period and temperature, respectively, are discussed in later sections.

The Cu/Pb modulated films present a metallic behaviour in the whole measured range of λ ; their resistivity decreases with decreasing temperature. It ranges from 11 to 21 $\mu\Omega \text{ cm}$ and from 3 to 13 $\mu\Omega \text{ cm}$ at room temperature and at 10 K, respectively, depending on the

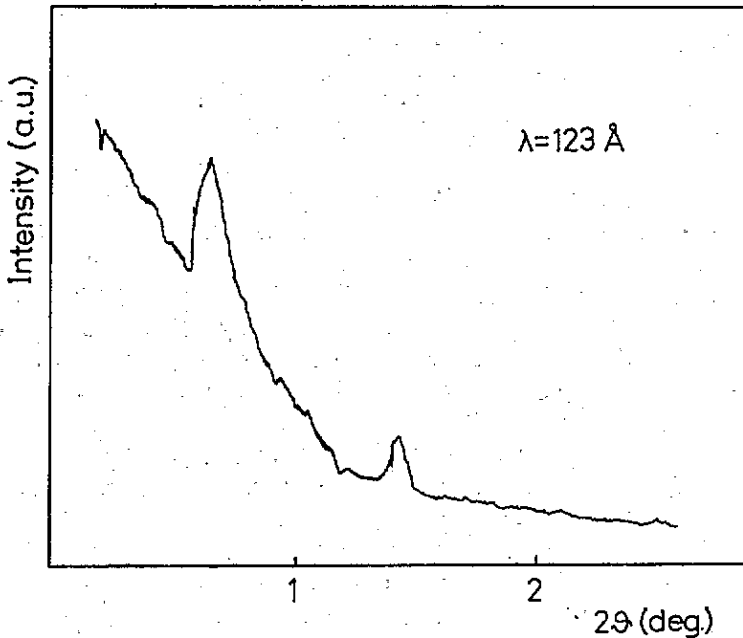


Figure 1. The small-angle x-ray diffraction pattern for modulated film with $\lambda = 123 \text{ \AA}$ (au, arbitrary units).

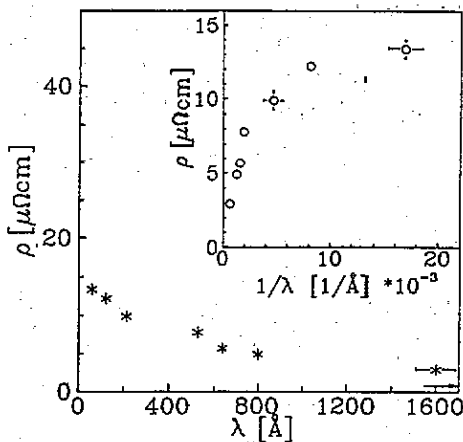


Figure 2. The dependence of the resistivity ρ on the modulation period λ at 10 K. The inset shows the resistivity ρ versus the reciprocal modulation period $1/\lambda$.

modulation period λ . The resistivity of modulated films seems to decrease asymptotically with increase in λ to the value indicated by the arrow in figure 2. To obtain reference values for use in the calculations following in the Gurvitch model, discussed below, we also prepared single films of Pb and of Cu with thicknesses of 5000 \AA each. The measured resistivities of these single layers at 10 K were $0.32 \mu\Omega \text{ cm}$ and $0.24 \mu\Omega \text{ cm}$ respectively,

and were taken as the bulk values. Such an assumption seems to be reasonable, because the influence of the crystalline and layered structure typical of the growth conditions in our vacuum system remains. Therefore the asymptotic value indicated in figure 2 is the resistivity of two resistors connected in parallel with resistivities equal to that obtained for single layers 5000 Å thick. However, even for large λ the resistivity still differs considerably from the asymptotic value. This is evidence of the shortening of the electron mean free path by the layer interfaces as well as by grain boundaries. We suppose that, because of the large lattice mismatch between Cu and Pb, the interfaces are strongly disordered and electrons are scattered diffusely. The clear influence of the modulated and polycrystalline structure on the electron mean free path is confirmed by the dependence of the resistivity on the reciprocal modulation period (inset in figure 2). One can distinguish between the regions by the different slopes of the plot. Usually in polycrystalline modulated films the grain diameters are proportional to the thickness of component layers. It is therefore reasonable to assume that both mechanisms—scattering at the interfaces and at the grain boundaries—contribute to the resistivity, resulting in a marked slope of the plot of resistivity versus $1/\lambda$ for longer λ . However, below a certain value of layer thickness, the grain size becomes constant, providing a smaller slope for shorter values of λ . Similar changes in ρ were observed for Mo/Cu modulated films [31].

To analyse the transport properties of component layers we followed the Gurvitch [30] model. This model allows us to calculate the resistivities of component layers and the electron mean free paths, which are not accessible in direct measurements. It treats the component layers of a modulated film as independent resistors connected in parallel. For this procedure, knowledge of the resistivities of modulated films only at room temperature and at low temperatures (at 10 K in our case) is necessary. For calculations we considered the general case, taking into account the saturation resistivity $\rho_{S(\text{Cu,Pb})}$. This value has an important meaning and is applied in the 'parallel-resistors' formula [32] for higher resistivities. The idea of saturation resistivity is based on the fact that the resistivity approaches a certain limiting value when the electron mean free path becomes comparable with the interatomic distance. The characteristic constant that relates the resistivity $\rho_{(\text{Cu,Pb})}$ of component layers with the electron mean free path $L_{(\text{Cu,Pb})}$ was estimated for free electrons:

$$C'_{(\text{Cu,Pb})} = \rho_{(\text{Cu,Pb})} L_{(\text{Cu,Pb})} = 178.1(A/\gamma z)^{2/3} \quad (1)$$

where A is the atomic mass, γ (g cm^{-3}) is the density and z is the valency. For Pb we assumed that $z = 4$. Using this valency we found that $C'_{(\text{Pb})}$ is equal to $492 \mu\Omega \text{ cm } \text{Å}$. The constant $C'_{(\text{Cu})}$ has already been estimated [30] as $815 \mu\Omega \text{ cm } \text{Å}$ from electron band calculations. Its value is 10% higher than that for free electrons. The values for the saturation resistivity

$$\rho_{S(\text{Cu,Pb})} = C'_{(\text{Cu,Pb})}/a_{n(\text{Cu,Pb})} \quad (2)$$

where $a_{n(\text{Cu,Pb})}$ is the nearest-neighbour distance in the lattice, were also evaluated as $140 \mu\Omega \text{ cm}$ and $332 \mu\Omega \text{ cm}$ for Pb and Cu, respectively. The nearest-neighbour distance $a_{n(\text{Cu,Pb})}$ was taken to be $a_{(\text{Cu,Pb})}/\sqrt{2}$ ($a_{(\text{Cu,Pb})}$ is the lattice constant) because both Cu and Pb have a FCC structure.

The dependence of the electron mean free path (for the general case, denoted as L) on the thickness d of component layers at 10 K is presented in figure 3. The value of the electron mean free path is calculated directly from equation (1). In both Pb and Cu layers, L is a linear function of d . Such a dependence is also evidence of the strong influence of

the modulated film structure, mentioned earlier, on its transport properties. For the region of short d , the question arises of whether the electron mean free path indeed exceeds the thickness of component layers. Also, in Ta/Nb superlattices [15] a similar phenomenon was observed. The electron mean free path exceeded the component layer thickness by several times in Nb/Ta films, much more than in our films. Because of the large lattice mismatch, interfaces are disordered in Cu/Pb films, contrary to the Nb/Ta superlattices, and we believe that electrons do not pass interfaces without scattering and the electron mean free path is rather limited by the component layer thickness.

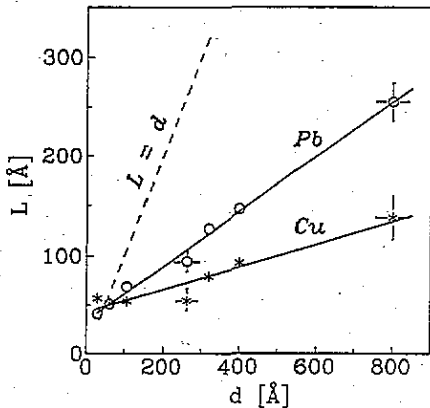


Figure 3. The electron mean free path L in the component layers Cu and Pb of a modulated film at 10 K.

3.2. Critical fields

The Cu/Pb films in the whole region of modulation periods become superconducting below a certain temperature. In this section we present the results of measurements of the critical fields versus temperature. The critical magnetic fields were measured in two configurations: parallel and perpendicular to the plane of the samples.

From the Ginzburg–Landau theory for anisotropic superconductors developed by Lawrence and Doniach [33], the critical fields are expressed in terms of the parallel coherence length ξ_{\parallel} and perpendicular coherence length ξ_{\perp} as follows:

$$H_{C_{\parallel}} = \Phi_0 / 2\pi \xi_{\parallel} \xi_{\perp} \quad (3)$$

$$H_{C_{\perp}} = \Phi_0 / 2\pi \xi_{\parallel}^2 \quad (4)$$

where Φ_0 is the flux quantum. In general, this theory predicts a crossover from a two-dimensional (2D) to a three-dimensional (3D) behaviour when ξ_{\perp} goes from $\xi_{\perp} \ll d_N$ to $\xi_{\perp} \gg d_N$ where d_N is the thickness of a normal metal component layer. In other words, the parabolic dependence of $H_{C_{\perp}}$ on temperature is evidence of 2D superconductivity whereas a linear dependence is evidence of 3D superconductivity. Following equations (3) and (4), one can evaluate the parallel coherence length ξ_{\parallel} and perpendicular coherence length ξ_{\perp} .

The dependences of the critical magnetic fields $H_{C_{\parallel}}$ and $H_{C_{\perp}}$ on temperature are presented in figure 4 for samples with modulation periods of 640, 790 and 1620 Å. The perpendicular critical fields $H_{C_{\perp}}$ are nearly linear versus temperature for all films, showing a slight saturation at low temperatures. However, the parallel critical fields are strongly influenced by the modulation period. The $H_{C_{\parallel}}$ for the sample with $\lambda = 640$ Å (figure 4(a))

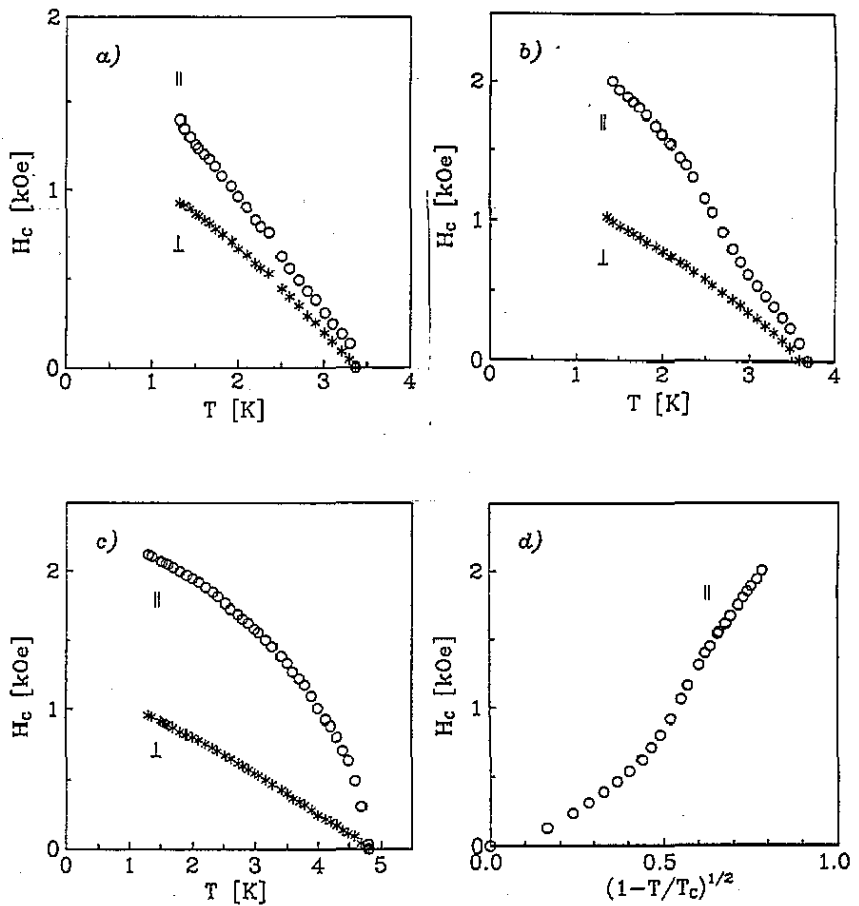


Figure 4. The dependence of the parallel critical field $H_{c||}$ (O) and the perpendicular critical field $H_{c\perp}$ (*) on temperature for Cu/Pb films with various modulation periods: (a) 640 Å; (b) 790 Å; (c) 1620 Å. (d) shows a parallel critical field $H_{c||}$ versus $(1 - T/T_c)^{1/2}$ for the film with $\lambda = 790$ Å.

(and for the other samples with shorter λ also measured but not presented in the figure) changes linearly with temperature. In these films the calculated perpendicular coherence length exceeds the thickness of the component layers in the whole range of temperatures, as expected for 3D superconductors. For the sample with a modulation period of 790 Å, the relation of $H_{c||}$ to temperature is more complex; at a temperature of 2.6 K the parallel critical field abruptly increases. Below this temperature the plot of $H_{c||}$ versus temperature has a parabolic shape. The parallel critical field as a function of $(1 - T/T_c)^{1/2}$ is shown in figure 4(d). The distinctly visible linear part of this plot corresponds to the parabolic dependence of $H_{c||}$ on temperature. In this sample the perpendicular coherence length takes the value of 286 Å at 1.3 K and increases with increasing temperature, reaching 400 Å at a temperature of 2.6 K (figure 5). It is interesting to note that at this temperature the dependence of critical parallel field changes from parabolic to linear. In agreement with theoretical expectations the dimensional 2D \rightarrow 3D crossover occurs therefore when $\xi_{\perp} \simeq d_N$. At a temperature higher than 2.6 K the perpendicular coherence length is larger than the thickness of the component layers. It involves the coupling of Pb component layers and a

3D type of superconductivity. In the sample with $\lambda = 1620 \text{ \AA}$ the parallel critical field is a parabolic function in the whole range of temperatures. The perpendicular coherence length in this film is slightly smaller than the thickness of the separating Cu component layers. Therefore Pb layers are decoupled and behave like a 2D superconductor.

Another parameter r of fundamental importance, which characterizes the strength of the two-dimensionality of the superconductors, is defined [34] by the following equation:

$$r = (4/\pi) (2\xi_{\perp}/\lambda)^2. \quad (5)$$

Roughly speaking $r < 1$ when the type of superconductivity is 2D whereas $r > 1$ for the 3D type. The calculated values of the parameter r at 1.3 K are 119, 10, 1.7 and 0.12 for the films with modulation periods of 59 \AA , 123 \AA , 640 \AA and 1620 \AA , respectively, confirming the above-mentioned criterion.

In all samples with a 3D type of superconductivity the linear dependence of $H_{c\parallel}$ on temperature deviates near T_c . The plot of $H_{c\parallel}$ versus T is concave in this region. It seems that none of the mechanisms discussed by Uher [35] (dimensional crossover, proximity effect, spin-orbit scattering and localization) is responsible for this effect. Moreover, these mechanisms should be active in the whole region of temperatures and not only near T_c . The existence of the above-mentioned deviation only near T_c suggests that the total thickness of the modulated film limits the perpendicular coherence length ξ_{\perp} . In accordance with theory the coherence length diverges to infinity when the temperature approaches T_c . At $t \simeq 0.95$ ($t = T/T_c$ is the reduced temperature) the perpendicular coherence length takes the value of approximately 5000 \AA in our samples and is therefore comparable with the total thickness of modulated films. Thus in this region of temperatures the modulated films as a whole behave like a 2D-type superconductor.

At the end of this section we try to estimate the density N of electronic states at the Fermi level in Cu/Pb modulated films on the basis of the perpendicular critical field dependence on temperature. For this purpose we apply the relationship between $H_{c\perp}$ and N in the following form [36]:

$$\mu_0(dH_{c\perp}/dT)_{T_c} = (8k_B/\pi)epN \quad (6)$$

where ρ is the resistivity of modulated film. In inhomogeneous materials such as modulated films, N is an average value of two constituents. Because the perpendicular critical field depends linearly on temperature, we assumed that

$$(dH_{c\perp}/dT)_{T_c} = (dH_{c\perp}/dT). \quad (7)$$

The calculated results are presented in figure 6. The highest value of N ($1.63 \times 10^{47} \text{ J}^{-1} \text{ m}^{-3}$) was obtained for the film with the largest modulation period. It exceeds the value of N for both Cu and Pb ($0.78 \times 10^{47} \text{ J}^{-1} \text{ m}^{-3}$ and $1.37 \times 10^{47} \text{ J}^{-1} \text{ m}^{-3}$ respectively [37]). The density of states decreases with decreasing λ , but this drop is less than 27%. The influence of N variation on critical temperature in Cu/Pb modulated films is analysed in the next section. For this purpose it was assumed that the density of states versus thickness of the component layer in Cu/Pb films was described by the equation

$$N = 0.140 \ln d + 0.668 \quad (8)$$

evaluated by the least-squares method, where d is in angströms and N in units of $10^{47} \text{ J}^{-1} \text{ m}^{-3}$.

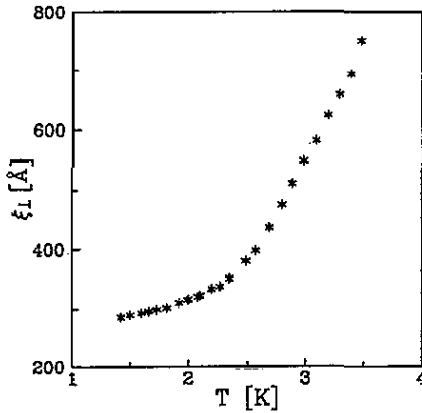


Figure 5. The dependence of the calculated perpendicular coherence length ξ_{\perp} on temperature for the sample with $\lambda = 790 \text{ \AA}$.

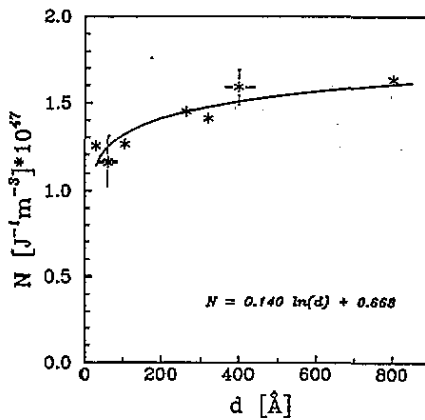


Figure 6. Calculated electron density N of states at the Fermi level as a function of the thickness d of component layers.

3.3. Critical temperature

The measured dependence of the critical temperature T_c on the thickness of component layers d is presented in figure 7 (asterisks). This temperature increases from 2.22 K for $d = 29 \text{ \AA}$ to 4.81 K for $d = 810 \text{ \AA}$, approaching asymptotically the value of T_c in bulk Pb (7.19 K).

We used the de Gennes–Werthamer model [38–40] to analyse this dependence. This model assumes that the superconductor energy gap is a position-dependent function Δ , which satisfies appropriate boundary conditions; $(1/\Delta)(d\Delta/dx)$ is continuous at the interfaces and $d\Delta/dx = 0$ at the metal–vacuum (substrate) surface. This problem is equivalent mathematically to the simple problem of the energy levels of a particle in a one-dimensional square potential well. As a result the following relations in the superconductor and normal metal are derived:

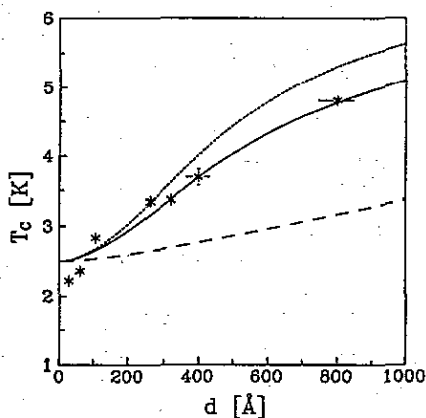


Figure 7. Experimental and theoretical dependences of the critical temperature on the thickness d of component layers (explanations in the text.)

$$\ln(T_{cs}/T_c) = \chi(\xi_s^2 k_s^2) \tag{9}$$

$$\ln(T_c/T_{cn}) = -\chi(-\xi_n^2 k_n^2) \tag{10}$$

$$[N\xi^2 k \tan(kd)]_s = [N\xi^2 k \tanh(kd)]_n \tag{11}$$

$$\xi_{s,n}^2 = \pi\hbar k_B / 6T_c e^2 (\rho\gamma)_{s,n} \tag{12}$$

$$\chi(z) = \psi(\frac{1}{2} + \frac{1}{2}z) - \psi(\frac{1}{2}) \tag{13}$$

where T_c , T_{cs} and T_{cn} are the critical temperatures of the modulated film, bulk superconductor and the normal metal, respectively, ξ is the effective coherence length, d is the thickness of component layers, ρ is the resistivity of component layers, γ is the coefficient of the electron specific heat in the normal state and ψ is the digamma function. Symbols with the subscripts s refer to the superconductor, whereas those with the subscript n refer to the normal metal.

To solve this problem we assumed as a first step that the density of states at the Fermi level was proportional to γ and was equal to the bulk value. Thus the values of γ were taken as $124 \text{ J m}^{-3} \text{ K}^{-2}$ and $163 \text{ J m}^{-3} \text{ K}^{-2}$ for Cu and Pb, respectively [41]. Because Cu is the normal metal down to the lowest measured temperatures, we took $T_{cn} = 0 \text{ K}$. In this limit, $\ln(T_c/T_{cn}) \rightarrow \infty$ and $k_n \rightarrow 1/\xi_n$, to satisfy equation (10). Moreover, we also assumed that T_{cs} did not depend on the thickness of the Pb component layer and was equal to the bulk value of Pb (7.19 K). Also the function $\chi(z)$ was replaced by the simpler expression [39]

$$\chi(z) = \begin{cases} \ln[1 + (\frac{1}{4}\pi^2 z)] & \text{for } z \geq 0 \\ \frac{1}{4}\pi^2 \ln(1+z) & \text{for } z < 0. \end{cases} \tag{14}$$

The assumptions discussed above provide the solution of the equation set (9)–(13) in the following form:

$$\begin{aligned} & (\gamma_n/\rho_n)^{1/2} \tanh[d_n(6T_c e^2 \gamma_n \rho_n / \pi \hbar k_B)^{1/2}] \\ & = (2/\pi)(\gamma_s/\rho_s)^{1/2} (T_{cs}/T_c - 1)^{1/2} \\ & \quad \times \tan[2(d_s/\pi)(6T_c e^2 \gamma_s \rho_s / \pi \hbar k_B)^{1/2} (T_{cs}/T_c - 1)^{1/2}]. \end{aligned} \tag{15}$$

The resistivities $\rho_{s,n}$ of the component layers appearing in equation (15) were found from the Gurvitch model for a temperature of 10 K. We also took into account that the electron mean free path varies linearly with the thickness of component layer (figure 3).

The numerical solution (full curve in figure 7) of equation (15) fits the experimental points very well. Only for $d \rightarrow 0$ does the critical temperature approach 2.2 K whereas the theoretical predictions provide a value of 2.5 K. It should be emphasized that this theoretical fitting was obtained without any adjustable parameters such as the dependence of T_{cs} on d_s [11] or the existence of diffusively mixed interlayers [16].

To explain the differences between the experimental and theoretical results of the critical temperature for thin component layers ($d \rightarrow 0$), we calculate the critical temperature in the Cooper [42] limit. For samples with the shortest modulation period the criterion of the Cooper limit is fulfilled, because the perpendicular coherence length exceeds several times the component layer thickness. The effective electron-electron interaction in normal metal/superconductor modulated films is then expressed as

$$(NV)_{s/n} = (N_s^2 V_s d_s + N_n^2 V_n d_n) / (N_s d_s + N_n d_n). \quad (16)$$

For our Cu/Pb modulated films we took $V_n = 0$, $N_n = 0.125 \times 10^{23} \text{ eV}^{-1} \text{ cm}^{-3}$, $N_s = 0.22 \times 10^{23} \text{ eV}^{-1} \text{ cm}^{-3}$ and $V_s = 1.7 \times 10^{-23} \text{ eV cm}^3$ [37]. The calculated value of the critical temperature from the equation

$$T_c = (\Theta_D / 1.45) \exp(-1 / (NV)_{s/n}) \quad (17)$$

is equal to 2.24 K. The Debye temperature Θ_D for the modulated film was taken as the average value of the Debye temperatures for Cu and Pb equal to 324 K and 88 K, respectively [37]. The above estimated critical temperature on the basis of equations (16) and (17) agrees very well with the temperature which experimental points reach in the limit $d \rightarrow 0$. Also in this case we calculated the value of T_c without any adjustable parameters.

On the basis of measurements of perpendicular critical field versus temperature we suggested in the previous section that the density of electronic states at the Fermi level varied slightly with the thickness of component layers (equation (8)). Because in equation (11) the value N occurs, we calculated once again how this the variation in N might influence T_c . We assumed that the variation in the density of states was the same in both the Cu and the Pb component layers (both values $\gamma_{s,n}$ in equation (15) scale). The result is plotted in figure 7 (broken (short dashes) line). The change introduced into the density of states increases the calculated critical temperature. In the limit $d \rightarrow 0$, both theoretical solutions become similar. When the thickness of the component layers increases, higher density of states than that for bulk material results in the higher critical temperatures, with a poorer fitting of the experimental points. Thus, finally one should believe that the density N of states in the Cu/Pb layers is constant and equal to the bulk value. The previously discussed procedure of N estimation on the basis of H_{c1} dependence on temperature is oversimplified and we assume that the calculated values might contain an error.

It ought to be mentioned that the boundary conditions play a very important role in analysing the T_c of the modulated films. A detailed discussion of this problem has been presented by Broussard [43]. We also calculated the critical temperature for the boundary conditions as for an infinite multilayer (in equation (11), $d_{s,n}$ is replaced by $\frac{1}{2}d_{s,n}$). The evaluated result is plotted in figure 7 as a broken (long dashes) curve. It is clear that with such boundary conditions there is no agreement between theoretical model and experimental points. The calculated critical temperature is considerably lower than the measured value, especially for the larger thickness of component layers.

4. Conclusions

We analysed the normal-state resistivity, critical magnetic fields and critical temperature in Cu/Pb films with various modulation periods. The layered structure considerably shortens the electron mean free path. In the dependence of the parallel critical field on temperature we found the 2D-3D crossover of superconductivity. It appears when the perpendicular coherence length is equal to the thickness of Cu component layers, which separate superconducting layers of Pb. The dependence of the critical temperature on modulation period is very well described by the de Gennes-Werthamer model with boundary conditions as for a double layer without any adjustable parameters.

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