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Critical fields of weakly and strongly coupled superconducting multilayers

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Abstract. We have compared the parallel ($H_{c2\parallel}$) and perpendicular ($H_{c2\perp}$) upper critical field of superconducting multilayers with strong and weak interlayer coupling (Pb/Cu, Pb/Ge). The temperature dependence of $H_{c2\parallel}$ reflects the expected dimensionality of the multilayers and can be interpreted in terms of the Takahashi–Tachiki proximity coupling model. In contrast with the strongly coupled Pb/Cu multilayers, weakly coupled Pb/Ge multilayers exhibit a strong thickness dependence of $H_{c2\perp}$. The electronic diffusion coefficient in Pb/Ge, obtained from $H_{c2\perp}$ data, is not in agreement with the value calculated from the normal state resistivity. The disagreement reflects the highly inhomogeneous nature of the weakly coupled Pb/Ge multilayers.

1. Introduction

Artificially prepared multilayered structures offer an ideal testing ground for the study of physical phenomena operating at different characteristic length scales (see, for example, [1]). In superconductors, the temperature dependent ratio between the layer thickness t and the coherence length ξ, gives rise to remarkable properties. In particular, the dimensional crossover in the temperature dependence of the parallel critical field was theoretically predicted [2] and experimentally observed in a number of multilayers, e.g. Nb/Cu [3], V/Ag [4], Nb/Ge [5], Pb/Ge [6].

In this paper we study the influence of the interlayer coupling on the parallel ($H_{c2\parallel}$) and perpendicular ($H_{c2\perp}$) critical fields in Pb based superconducting multilayers. The coupling strength between the Pb layers can be varied by a suitable choice of the separating layer material. A normal metal (Cu) produces a strong proximity effect coupling between the superconducting Pb layers. For a semiconducting or insulating material (Ge), only a weak coupling via Josephson or one-electron tunnelling is possible. Nevertheless, our Pb/Ge multilayers also show a characteristic proximity coupling of the Pb layers with a thin surface layer of the neighbouring Ge layers. The choice of Cu and Ge enables us to compare the critical fields for strong and weak *proximity coupling*. The temperature dependence of $H_{c2\parallel}$ shows the expected behaviour as a function of

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interlayer coupling strength. We also find that the perpendicular critical field $H_{c2\perp}$ depends strongly on the electronic properties of the interface. The stronger flux pinning near the Pb/Ge interface considerably enhances the perpendicular critical field.

2. Experimental details

Pb based multilayers offer several advantages over other multilayer systems: the low melting temperature of Pb leads to an accurate control of the evaporation rate, and the critical temperature T_c of the Pb layers is fairly insensitive to the amount of impurities incorporated in the film. The thermodynamic phase diagram of Pb/Cu and Pb/Ge shows that these materials form no compounds or solid solutions, indicating that there is very little interdiffusion. The samples were made in a UHV chamber equipped with two electron beam guns [7]. The base pressure of the system is 2×10^{-9} Torr, and the pressure increased during evaporation up to 10^{-8} Torr. The evaporation rates of the two materials were kept constant using a quadrupole mass spectrometer. The multilayered structure was obtained by alternately interrupting one of the material beams using a shutter. Typical evaluation rates are 5 Å s^{-1} for Pb and Cu and 2 Å s^{-1} for Ge. In order to assure a uniform layer growth for the Pb layers, the samples with Pb layer thickness below 200 Å were evaporated onto liquid nitrogen cooled substrates (oxidised silicon wafers or sapphire). Samples with thicker Pb layers were evaporated on substrates held at room temperature. The layer thicknesses, as monitored with quartz crystals during evaporation, were calibrated using a Dektak profilometer. All multilayers consist of 10 bilayers. The top and bottom layers are always Cu or Ge, in order to decrease surface superconductivity effects. The four-point pattern (4.5 mm × 0.3 mm), needed for resistivity measurements, was defined photolithographically using a lift-off technique. The measurements were performed in a standard ^{4}He cryostat, equipped with a 7 T superconducting coil. The temperature was varied from 1.5 K to 10 K with a stability of a few mK. Both the critical temperature T_c and the critical field H_{c2} are defined as the midpoint values of the DC measured $R(T)$ and $R(H)$ transitions. Layer uniformity of the Pb/Cu samples with layer thicknesses above 150 Å was confirmed by Rutherford backscattering (RBS); the structural quality of the thinner Pb/Cu and Pb/Ge samples was extensively investigated by x-ray diffraction. Kinematic and dynamic modelling of the $\theta - 2\theta$ diffraction profiles of the Pb/Cu (crystalline/crystalline) and the Pb/Ge (crystalline/amorphous) multilayers indicate discrete and continuous thickness fluctuations of a few Å [8–10].

3. Results and discussion

3.1. Parallel critical fields

According to the Ginzburg–Landau theory, the parallel critical field near T_c of a three-dimensional (3D) anisotropic superconductor is given by:

$$H_{c2\parallel}(T) = \Phi_0/2\pi\xi_\parallel(T)\xi_\perp(T) \tag{1}$$

where Φ_0 is the superconducting flux quantum and $\xi_\parallel(\xi_\perp)$ are the coherence length parallel (perpendicular) to the layers. With $\xi(T) \propto (T_c - T)^{-1/2}$, (1) predicts a linear temperature dependence for $H_{c2\parallel}$.

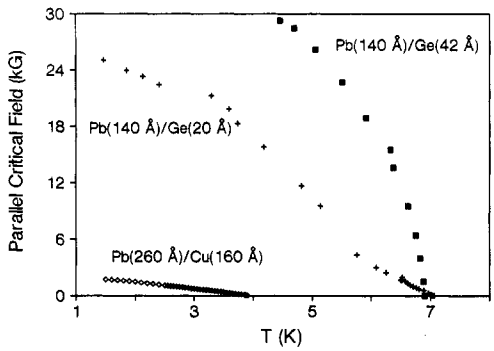

Figure 1. Temperature dependence of the parallel critical field for a Pb(260 Å)/Cu(160 Å) (◇), a Pb (140 Å)/Ge(20 Å) (+) and a Pb(140 Å)/Ge(42 Å) (■) multilayer.

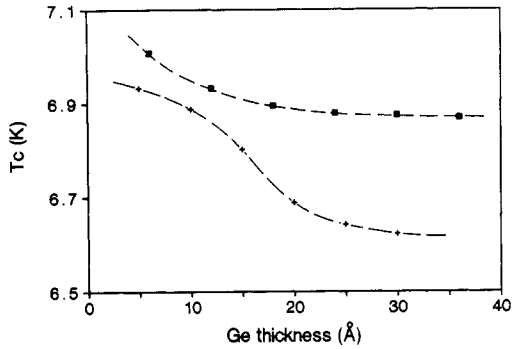

Figure 2. Critical temperature of two sets of Ge/Pb/Ge/Pb/Ge multilayers as a function of Ge thickness. The Pb thickness t_{Pb} for each set was kept constant (t_{Pb} = 140 Å (■) and t_{Pb} = 70 Å (+)). The dashed lines are a guide to the eye.

The parallel critical field for a thin superconducting slab of thickness $t \ll \xi_{\|}$, was calculated by Tinkham [11]:

$$H_{c2\|}(T) = \Phi_0\sqrt{12}/2\pi\xi_{\|}(T)t. \tag{2}$$

For uncoupled thin Pb layers the temperature dependence of $\xi_{\|}$ now implies a square-root temperature dependence of $H_{c2\|} \propto (T_c - T)^{1/2}$.

The linear temperature dependence of $H_{c2\|}(T)$ (figure 1) for the Pb/Cu multilayers with 200 Å $< t_{Pb} <$ 500 Å and $t_{Cu} \leqslant t_{Pb}$ clearly indicates a 3D coupled behaviour. For Pb/Ge multilayers with $t_{Ge} > 30$ Å, the square-root temperature dependence of $H_{c2\|}$ indicates that the Pb layers are essentially uncoupled. As the Ge thickness is decreased below 30 Å, dimensional crossover (3D-2D) occurs. The change from an uncoupled to a coupled behaviour is caused by the divergence of $\xi_{\perp}$ in the vicinity of T_c. When $\xi_{\perp}(T)$ becomes comparable to the multilayer period $\Lambda(\Lambda = t_{Pb} + t_{Ge})$, an effective superconducting coupling of the Pb layers is established [12]. For our Pb(140 Å)/Ge(20 Å) multilayer the 3D coupling is achieved for $T > 5.5$ K.

The fact that the uncoupled Pb(140 Å)/Ge(42 Å) multilayer has a slightly lower T_c than the coupled Pb(140 Å)/Ge(20 Å) multilayer, points to a possible proximity coupling between the Pb and Ge layers. In order to prove unambiguously the existence of this proximity effect, we prepared a series of Ge/Pb/Ge/Pb layers. The Pb layers were evaporated simultaneously for all the samples, in order to rule out changes in T_c due to Pb thickness fluctuations. The Ge thickness was varied by moving a shutter across the already deposited Pb layers. The Ge/Pb/Ge/Pb layers were evaporated onto a predefined lift-off structure of 2 cm × 1 cm containing six four-point patterns. All samples were covered with a 500 Å Ge protective layer. The critical temperature as a function of Ge thickness for two sets of double-layers is shown in figure 2. The initial decrease of T_c for small Ge thicknesses ($t_{Ge} < 30$ Å) and the saturation at larger Ge thicknesses ($t_{Ge} > 30$ Å) for a constant Pb thickness indicates that the Pb layers are proximity coupled with a thin Ge surface layer of about 15 Å. This weak proximity coupling is sufficient to establish a 3D coupled behaviour of $H_{c2\|}$ close to T_c for Ge thicknesses < 30 Å.

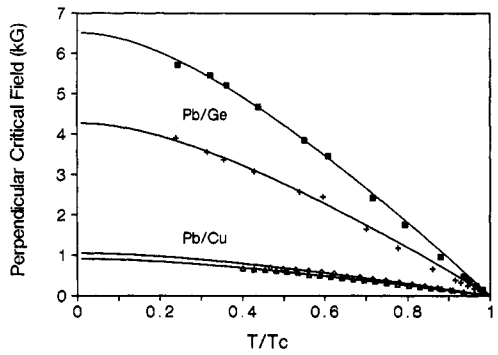

Figure 3. Perpendicular critical field as a function of reduced temperature T/T_c for two Pb/Ge multilayers (126 Å/30 Å (■) and 245 Å/30 Å (+)), and two Pb/Cu multilayers (200 Å/200 Å (◇) and 500 Å/500 Å (△)). The solid lines are a fit using the dirty-limit Maki-de Gennes model (equation (4)).

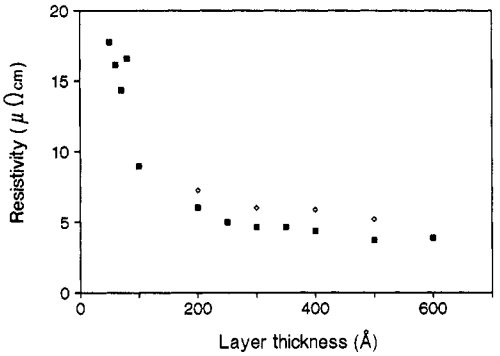

Figure 4. Multilayer resistivity for equal layer thickness Pb/Cu multilayers as a function of the individual layer thickness, determined from resistance measurements (■) and from the perpendicular critical field (◇) (equation (4)).

The dimensional crossover phenomenon observed in $H_{c2\parallel}(T)$ was quantitatively explained by Takahashi and Tachiki [2]. Their microscopic theory for proximity coupled multilayers takes into account the spatial variation of the density of states, the diffusion constant and the BCS electron–electron interaction. These quantities are assumed to be uniform inside the layers and should change discontinuously at the interfaces. The presence of the dimensional crossover is mainly caused by the difference in the density of states at the Fermi level in the superconducting (N_S) and normal material (N_N). When the ratio N_N/N_S approaches unity, e.g. for Pb/Cu multilayers $N_{Cu}/N_{Pb} = 0.72$, the model predicts a linear temperature dependence of $H_{c2\parallel}$. For superconductor/insulator or superconductor/semiconductor multilayers ($N_N/N_S = 0$), the theory predicts the 2D square-root dependence and much higher values of $H_{c2\parallel}$. This is indeed observed in Pb/Ge multilayers with $t_{Ge} \geqslant 30$ Å. However, in multilayers with $t_{Ge} < 30$ Å, the temperature at which the dimensional crossover occurs, is consistent with a value $N_{Ge}/N_{Pb} \simeq 0.05$. The finite value for the density of states in the Ge layers may be due to the amorphous structure [8] of the Ge. It is clearly a surface effect and also explains the appearance of a proximity effect (see figure 2) for very thin Ge layers.

3.2. *Perpendicular critical fields*

The Ginzburg–Landau formula for the perpendicular critical field of a bulk anisotropic superconductor is:

$$H_{c2\perp}(T) = \Phi_0/2\pi\xi_\parallel^2(T). \tag{3}$$

Figure 3 shows the temperature dependence of $H_{c2\perp}$ as a function of the reduced temperature T/T_c for two Pb/Cu multilayers (200 Å/200 Å and 500 Å/500 Å), and two Pb/Ge multilayers (126 Å/30 Å and 245 Å/30 Å). In all cases, $H_{c2\perp}$ is a linear function of temperature close to T_c, in agreement with equation (3). The perpendicular critical field for the anisotropic 3D Pb/Cu multilayers is small and almost independent of the Pb layer thickness, whereas $H_{c2\perp}$ for the uncoupled, 2D Pb/Ge multilayers is higher and

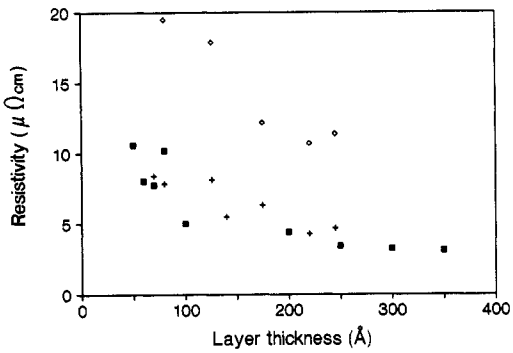

Figure 5. Resistivity per Pb layer calculated from the multilayer resistance of Pb/Cu (■), Pb/Ge (+) and from $H_{c2\perp}$ measurements of Pb/Ge (◇) using equation (4).

very sensitive to the Pb thickness. The increase of $H_{c2\perp}$ is probably due to a change in the electronic mean free path. For dirty 3D superconductors the influence of a reduced mean free path can be described using the Maki-de Gennes relation [13]:

$$\ln(T_c/T) = \psi(\tfrac{1}{2} + eDH_{c2\perp}/2\pi k_B T) - \psi(\tfrac{1}{2}) \tag{4}$$

where ψ is the digamma function and D is the diffusion constant. $D = (\pi^2 k_B^2)/(3e^2\gamma\rho)$, with γ the coefficient of the linear electronic specific heat and ρ the resistivity. The full curves in figure 3 have been calculated using equation (4). The proximity effect is taken into account by assigning to T_c the critical temperature of the multilayer. Introducing an extra pair-breaking parameter caused by the proximity effect and assuming $T_c = 7.2$ K, yielded very similar results.

The thickness independence of the $H_{c2\perp}$ values in Pb/Cu multilayers can be understood by the competition between the proximity effect and the enhanced flux pinning in dirty layers ($H_{c2\perp} \propto \rho$). In Pb/Ge multilayers, the weak proximity coupling affects T_c only slightly, and the thickness dependence is caused by the strong scattering at the Pb/Ge interface.

The resistivity $\rho = (\pi^2 k_B^2)/(3e^2\gamma D)$ was determined for several uncoupled Pb/Ge ($t_{Ge} \geqslant 30$ Å) and coupled Pb/Cu multilayers, using equation (4). Figure 4 shows the low temperature total resistivity of equal layer thickness Pb/Cu multilayers, as a function of the individual layer thickness, calculated from the resistance (at 4.2 K) and $H_{c2\perp}$ measurements. Since all layers have the same thickness, we took into account an average $\gamma = (\gamma_{Pb} + \gamma_{Cu})/2$. For γ_{Pb} and γ_{Cu} we used the experimentally determined values [14]. The good agreement between the ρ values as a function of layer thickness for both calculation methods, confirms the homogeneous nature of the coupled Pb/Cu multilayers (which means a relatively small perturbation of the order parameter across the Pb/Cu interface).

For the uncoupled Pb/Ge multilayers, equation (4) enables us to calculate the resistivity of one single Pb film in proximity with a thin Ge surface layer of about 15 Å. Since $\gamma_{Ge}/\gamma_{Pb} = N_{Ge}/N_{Pb} \ll 1$, we used the γ_{Pb} to calculate the resistivity for the Pb in proximity with Ge. The ρ value is much higher than the resistivity obtained for a single Pb film, either measured directly in Pb/Ge multilayers (at 7.5 K), or extracted from the Pb/Cu multilayer resistivity using Gurvitch's parallel resistors model [15] as shown in figure 5. This discrepancy results from the inhomogeneous nature of the uncoupled Pb/Ge multilayers. The normal transport resistivity is not affected by the high resistivity of the Ge films. However, $H_{c2\perp}$ measurements sample the region with the highest resistivity.

The higher flux pinning near the Pb/Ge boundary leads to an increased $H_{c2\perp}$. This discrepancy is naturally not present in the Pb/Cu multilayers. The high resistivity of the Ge layers and the active part they play in the superconducting transport explains the higher values of the perpendicular critical fields of Pb/Ge multilayers in comparison with the Pb/Cu multilayers.

In summary, a detailed analysis of the critical fields of Pb/Cu and Pb/Ge multilayers showed that $H_{c2\parallel}(T)$ is determined by the dimensionality of the multilayer while $H_{c2\perp}(T)$ strongly depends on the interface properties. In strongly coupled Pb/Cu multilayers, the competition between the proximity effect and the disorder gives rise to a thickness independent $H_{c2\perp}$. The $H_{c2\perp}$ value for the weakly coupled Pb/Ge multilayers is mainly determined by the highly resistive interface layer.

Acknowledgments

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