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Crossover in the critical field of Pb/Ge multilayers: From single-film to coupled behavior

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A new type of crossover is observed in the temperature dependence of the parallel critical field of superconductor-insulator multilayers. This transition from a two-dimensional single-film behavior towards a two-dimensional coupled behavior results from the limited number of bilayers in the multilayer system.

An understanding of the nature of dimensional transition in superconductors has been the subject of great theoretical[1] and experimental[2] interest. Normal and superconducting transport and magnetic properties have been extensively studied in single layers with thicknesses smaller than the characteristic electronic mean free path and (or) the superconducting coherence length.[3] Recent developments in fabrication techniques have given an important impetus to the study of dimensional transitions in artificially prepared multilayers. In this framework, proximity and Josephson coupling between thin superconducting layers across normal or insulating layers has been studied. A strong coupling between the superconducting layers occurs when the perpendicular coherence length $\xi_\perp$ is comparable to the multilayer period Λ (equal to the sum of the superconducting and normal layer thickness).[4] Near the critical temperature T_c, where $\xi_\perp$ diverges, this coupling produces a change of the temperature dependence of the parallel critical field $H_{c2\parallel}$ from a square-root-like two-dimensional (2D) behavior at lower temperature to a linear (3D) behavior at higher temperature. This dimensional crossover has been observed in a large number of multilayered systems,[5-8] and the crossover temperature largely depends on the difference in density of states of the constituent materials of the multilayer.[9]

We investigated the parallel critical field of Pb/Ge multilayers consisting only of two Pb/Ge bilayers. One single Pb layer has a thickness smaller than the superconducting coherence length $\xi_\parallel$ in the plane of the Pb layer ($\xi_\parallel \simeq 250$ Å at $T=0$ K), resulting in a 2D square-root-like temperature dependence of $H_{c2\parallel}$. Because of the limited number of bilayers, the total multilayer also has a thickness smaller than $\xi_\parallel$. This causes also a 2D temperature dependence of $H_{c2\parallel}$ for the coupled multilayer (in case of small Ge thickness), but with a different amplitude. For sufficiently large Ge thickness, the weak interlayer coupling induces a 2D, single-film behavior at lower temperature, which changes towards a 2D coupled behavior near T_c. This remarkable 2D-2D crossover in superconducting multilayers confirms the importance of the ratio between total multilayer thickness and the *parallel* coherence length $\xi_\parallel$ for the determination of the overall dimensionality of the coupled multilayer (when $\xi_\perp > \Lambda$).

The Pb/Ge multilayer samples were prepared in an UHV chamber equipped with two electron beam guns.[10] The base pressure of the system is 2×10^{-9} Torr, and the pressure did not exceed 10^{-8} Torr during the evaporation. The evaporation rates of the two materials are controlled by a quadrupole mass spectrometer and are respectively 5 Å/s for Pb and 1 Å/s for Ge. In order to ensure a uniform layer growth for the Pb, the samples were evaporated onto liquid nitrogen cooled substrates (oxidized silicon wafers or sapphire). This low substrate temperature leads to an amorphous structure of the Ge films. The layer thicknesses, as monitored with quartz crystals during evaporation, were calibrated using a Dektak profilometer. The Pb/Ge multilayer samples discussed in this paper consist of a Ge/Pb/Ge/Pb/Ge sequence, where the first two Ge layers have the same thickness, denoted by $d_{\rm Ge}$, and the top Ge layer is a 500-Å-protective layer. All samples were evaporated onto a photolithographically defined lift-off structure containing six four point patterns of 4.5×0.3 mm^2. We prepared two sets of six samples. The first set has a Pb thickness $d_{\rm Pb}=70$ Å while the second set has $d_{\rm Pb}=140$ Å. The thickness $d_{\rm Ge}$ of the first two Ge layers in each set is varied between 5 and 40 Å. The Pb layers were evaporated simultaneously for all samples in one set, in order to rule out spurious changes in structure, composition, film thickness, etc., which may influence T_c. The different Ge thicknesses are obtained by moving a shutter across the substrate during evaporation. The low-temperature measurements were performed in a standard ^{4}He cryostat with a 7 T superconducting coil. The temperature could be varied between 1.5 and 10 K with a stability of a few mK. The critical temperature T_c and the critical field H_{c2} are defined as the midpoint values of the dc four probes measured $R(T)$ and $R(H)$ transitions, respectively.

The zero-field resistance versus temperature transition widths (10%–90%) were always smaller than 0.03 K, indicating the homogeneous structure of the films. The critical temperature T_c of the two sets of Pb/Ge multilayers slightly decreases with increasing Ge thickness, similar to the observation in Nb/Ge multilayers.[11] A full discussion of this thickness dependence of T_c and its possible origin will be the subject of a future publication. At this point we only note that both Josephson tunneling through a Ge layer and proximity coupling of Pb with Ge cannot be discarded. Both mechanisms can cause a superconducting coupling of the individual Pb layers across a Ge layer. This coupling is revealed by measuring the parallel upper critical field. Figure 1 shows the upper critical fields parallel to the layers ($H_{c2\parallel}$) and perpendicular to the layers ($H_{c2\perp}$) as a function of the reduced temperature T/T_c for the set of Pb/Ge multilayers with $d_{\mathrm{Pb}}=70$ Å and different d_{Ge}. The temperature dependence of $H_{c2\perp}$ is linear as expected and does not change appreciably with the Ge thickness. Using the Ginzburg-Landau relation $H_{c2\perp}(T)=\Phi_0/2\pi\xi_{\parallel}^2(T)$, with Φ_0 the superconducting flux quantum, we extract the parallel coherence length $\xi_{\parallel}(T=0)\simeq 220$ Å for all samples. Since $\xi_{\parallel} > d_{\mathrm{Pb}}$, the individual Pb films are 2D, provided the structure of the Pb films is isotropic. Since $\xi_{\parallel}$ is also larger than the sum of the two Pb/Ge bilayer thicknesses ($\xi_{\parallel} > 2\Lambda$), we also expect a 2D behavior for the coupled multilayer (if $\xi_{\perp} > \Lambda$). The 2D nature of a superconductor is revealed by the temperature dependence of $H_{c2\parallel}$, as calculated by Tinkham:

$$H_{c2\parallel}=\frac{\Phi_0}{2\pi}\frac{\sqrt{12}}{d_{\mathrm{eff}}\xi_{\parallel}(T)}\,, \tag{1}$$

with d_{eff} the effective superconductor thickness. Since $\xi_{\parallel}$ varies as $(T_c-T)^{-1/2}$, $H_{c2\parallel}$ for a 2D superconductor has a square-root-like temperature dependence: $H_{c2\parallel}\propto(T_c-T)^{1/2}$.

As shown in Fig. 1, $H_{c2\parallel}\propto(T_c-T)^{1/2}$ for the multilayer with $d_{\mathrm{Ge}}=5$ Å. The amplitude of $H_{c2\parallel}(T)$ coincides exactly with the measured critical field of a 140 Å single Pb film, sandwiched between Ge (not shown). Indeed, the effective superconducting layer thickness $d_{\mathrm{eff}}\simeq 130$ Å as calculated from Eq. (1), is approximately equal to $2\Lambda=150$ Å indicating that the two Pb layers are coupled through the Ge layer. Since 2Λ is smaller than $\xi_{\parallel}(0)$, the coupled multilayer behaves as a 2D superconductor. Increasing the Ge thickness initially leads to a decrease of $H_{c2\parallel}$ for $d_{\mathrm{Ge}}=15$ Å, until for $d_{\mathrm{Ge}}=20$ Å a remarkable upturn of $H_{c2\parallel}$ occurs at lower temperature. This enhancement indicates a crossover from the parallel critical field of the 2D coupled multilayer close to T_c, to the parallel critical field of a 2D, 70-Å-Pb film at lower temperature. It should be noted that $H_{c2\parallel}$ of the $d_{\mathrm{Ge}}=20$ Å and the $d_{\mathrm{Ge}}=25$ Å multilayers has a 2D square-root-like temperature dependence below as well as above the crossover temperature. This 2D-2D crossover is characterized by a transition from a 2D single film behavior towards a 2D coupled behavior. A further increase of d_{Ge} drives the critical field towards the measured $H_{c2\parallel}$ value of a 70-Å-Pb film sandwiched between two Ge layers (dashed line in Fig. 1).

Figure 2 shows detailed measurements of the resistive transition of a similar two-bilayer Pb/Ge multilayer with $d_{\mathrm{Pb}}=80$ Å and $d_{\mathrm{Ge}}=25$ Å. The resistance of the multilayer is plotted as a function of parallel magnetic field for several values of T/T_c in the vicinity of the 2D-2D crossover. When $T/T_c=0.985$, the transition of the coupled multilayer is sharp and is comparable to the resistive transition of a 160-Å-Pb film, sandwiched between Ge. For $T/T_c=0.971$ and 0.966, the 2D-2D crossover is seen by a decrease of the resistance with increasing parallel magnetic field. This decrease is caused by a sudden increase of $H_{c2\parallel}$ upon decoupling of the Pb layers. When $T/T_c<0.944$, the transition corresponds to the resistive transition of a 80-Å-Pb film, sandwiched between Ge. The resistance versus parallel field behavior shows the importance of the parallel magnetic field in the decoupling mechanism. The role of a parallel field in the usual 2D-

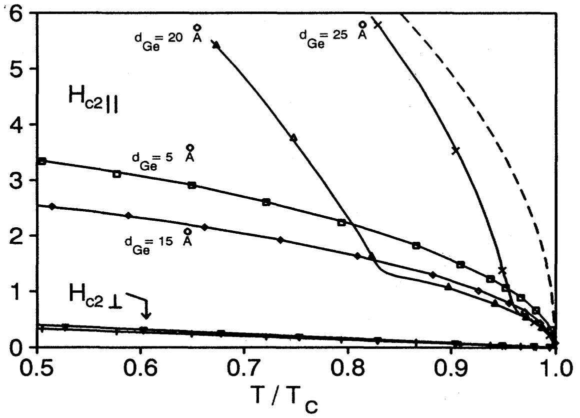

FIG. 1. Parallel critical field of four samples from the $d_{\mathrm{Pb}}=70$ Å multilayer set with differing Ge thickness [5 Å (□); 15 Å (◇); 20 Å (△); 25 Å (×)]. The perpendicular critical field $H_{c2\perp}$ is shown for the $d_{\mathrm{Ge}}=5$-Å sample (+) and the $d_{\mathrm{Ge}}=25$-Å sample (▽). The dashed line represents the parallel critical field of a 70-Å-single Pb film sandwiched between thick Ge. The solid lines are a guide to the eye. The 2D-2D crossover is clearly seen in the samples with $d_{\mathrm{Ge}}=20$ Å (△) and $d_{\mathrm{Ge}}=25$ Å (×).

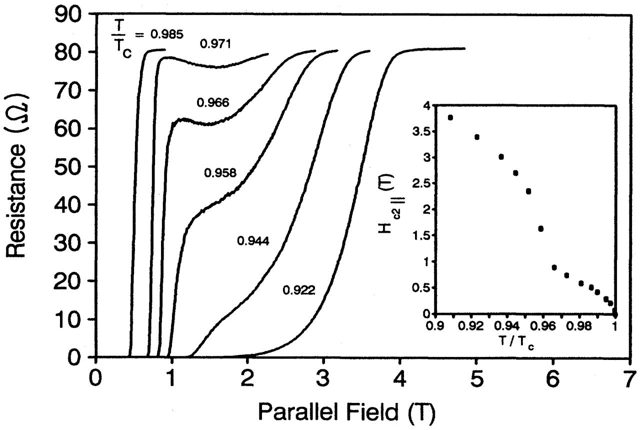

FIG. 2. Resistance versus parallel magnetic field of a Pb(80 Å)/Ge(25 Å) multilayer consisting of two bilayers, for several values of T/T_c. The 2D-2D crossover is seen by a decrease of the multilayer resistance with increasing field. The inset shows $H_{c2\parallel}$ as a function of reduced temperature, determined by the midpoint values of the resistive transition.

3D crossover of Pb/Ge multilayers was recently emphasized by measurements of the fluctuation conductivity.[12] The inset of Fig. 2 presents $H_{c2\parallel}$ of the Pb(80 Å)/Ge(25 Å) multilayer, as determined from the midpoint values of the resistive transition. The square-root-like, and thus 2D temperature dependence of $H_{c2\parallel}$ is clearly recognized below as well as above the crossover temperature.

The role of the number of bilayers in the problem of dimensionality can be demonstrated when we analyze the temperature dependence of $H_{c2\parallel}$. Figure 3 shows $\ln H_{c2\parallel}$ vs $\ln(1-T/T_c)$ for two multilayers with the same Ge thickness ($d_{\mathrm{Ge}}=20$ Å) and the same Pb thickness ($d_{\mathrm{Pb}}=140$ Å), but a different number of bilayers. For a Pb(140 Å)/Ge(20 Å) multilayer with only two bilayers $H_{c2\parallel}\propto(1-T/T_c)^{0.57}$ close to T_c indicating the 2D nature. Since the superconducting effective layer thickness ($d_{\mathrm{eff}}\simeq 250$ Å) is of the same magnitude as the multilayer thickness ($2\Lambda=320$ Å), the two Pb layers are coupled. At lower temperature, decoupling occurs and the critical field tends toward the $H_{c2\parallel}$ of the single 140-Å-Pb film (dotted line in Fig. 3). On the other hand, the temperature dependence of $H_{c2\parallel}$ of a Pb(140 Å)/Ge(20 Å) multilayer with *ten* bilayers is completely different. At low temperature, the behavior is identical to the decoupled Pb(140 Å)/Ge(20 Å) two-bilayer sample. When coupling occurs (close to T_c), $H_{c2\parallel}\propto(1-T/T_c)^{1.15}$ indicating that the coupled ten-bilayer multilayer displays a 3D behavior. The different dimensionality for the coupled ten-bilayer multilayer compared to the two-bilayer multilayer, confirms the importance of the number of coupled layers to determine the effective dimensionality of the system.

In summary, we considered a multilayered system consisting of N two-dimensional superconducting layers ($d_{\mathrm{Pb}}<\xi_\parallel$), separated by Ge. An effective coupling of the superconducting layers can be established, depending on the Ge thickness. The dimensionality of the coupled multilayer is still two-dimensional if $N\Lambda<\xi_\parallel$. In our experiment we limited the number of superconducting layers ($N=2$). One may also decrease the superconducting layer thickness to observe a 2D coupled behavior even with a larger number of bilayers (N). In the limit of infinitely thin superconducting atomic planes coupled through nonsuperconducting layers, a 2D behavior may still be possible for very large thicknesses.

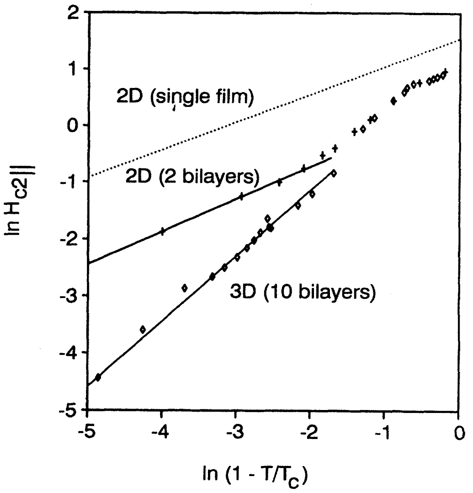

FIG. 3. Power-law temperature dependence of the critical field $H_{c2\parallel}$ showing the 2D-3D transition [10×Pb(140 Å)/Ge(20 Å); ◇] and the 2D-2D transition [2×Pb(140 Å)/Ge(20 Å); +]. The dotted line represents the single film behavior.

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