Transition in the Flux Lattice of Artificially Layered Superconductors

D. Neerinck, K. Temst, M. Baert, E. Osquiguil, C. Van Haesendonck, and Y. Bruynseraede
Laboratorium voor Vaste Stof-Fysika en Magnetisme, Katholieke Universiteit Leuven, B-3001 Leuven, Belgium

A. Gilabert
Laboratoire de Physique de la Matière Condensée, Université de Nice, F-06034 Nice CEDEX, France

Ivan K. Schuller
Physics Department, University of California, San Diego, La Jolla, California 92093-0319
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We report a novel magnetic-field dependence of the critical-current density J_c in artificially grown Ge/Pb superlattices. In these layered structures J_c exhibits an unexpected minimum as a function of *perpendicular* applied field, unlike any ever observed in other superconducting systems. The systematic evolution of this minimum as a function of layer thicknesses, temperature, and pinning strength is related to either flux-lattice decoupling or melting.

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The behavior of the flux structure of layered superconductors has been of major interest in recent years, especially in its relation to the properties of high-temperature superconducting oxides [1,2]. The H-T phase diagram of a number of superconductors [3-7] shows, close to $H_{c2}(T)$, a novel phase boundary above which the magnetization of the superconductor is reversible and below which irreversibility sets in. The origin of this phase boundary has also been the subject of intense theoretical interest, especially including a quasi Thouless–de Almeida line [4], vortex depinning by thermal fluctuations [3], or melting of the flux-line lattice [8-12].

Many of these theories rely on the layered nature of the superconductor and deal with a competition of pinning of the vortex lattice in the individual layers or the multilayered stack as a whole. Surprisingly, the low-field region $H \simeq H_{c1}$ has been much less explored. A very interesting prediction [8,13] in this region for systems with low pinning is the presence of an additional reentrant vortex fluid phase above H_{c1}. The flux lattice present at high fields melts at low fields because the vortex-vortex interaction in this region is exponentially weak and therefore the flux lattice is highly unstable to thermal fluctuations.

Within this context artificially layered superconductors offer a major advantage over high-T_c oxide superconductors since it is possible to vary almost at will the superconductor layer thicknesses, the superconducting properties of these layers, and the interlayer coupling (and thus the anisotropy). We show here that Ge/Pb multilayers [14] exhibit an unusual H-T phase diagram for fields $H \gtrsim H_{c1}$ applied *perpendicular* to the multilayer planes. This phase boundary is signaled by a very unusual field dependence of the critical current J_c and is strongly dependent on the multilayered nature of the material, interlayer spacing, pinning, and temperature. These changes [13,15] in the flux-line lattice are thermally driven and introduce a low-field $H(T)$ line in the phase diagram which is possibly connected to the low-field melting transition in the vortex structure [13] or magnetic decoupling [16-18] of the layers.

High-quality multilayered Ge/Pb superconductors have been prepared, characterized, and studied for a number of years in our laboratories. The samples are evaporated in a molecular-beam-epitaxy apparatus on nitrogen-cooled SiO_2 substrates using electron-beam guns controlled by a mass spectrometer [19]. The structure of these superlattices has been the subject of intense x-ray-diffraction studies and simulations showing that the structure is well layered, with negligible interdiffusion and an interfacial roughness of less than 2 Å discrete roughness on the crystalline Pb and less than 2 Å continuous roughness on the amorphous Ge [20]. In addition, the fact that Pb and Ge do not form any intermetallic alloys [21] favors the growth of a highly segregated layered material [22]. The conclusions presented here are based on the investigation of more than 25 samples, which have a $[\mathrm{Ge/Pb}]_n\mathrm{Ge}$ structure, where n denotes the number of bilayers and the final Ge film is always a 500-Å protective layer.

The critical current is derived from I-V curves using four-probe dc resistivity measurements. The four point structures (4.5 mm × 0.3 mm) are obtained by a lift-off technique using electron-beam lithography. The voltage criterion used for the determination of the critical currents is 4.4 μV/cm, although other criteria give similar results. The critical current density is calculated using the total Pb cross section only, since the Ge layers are not superconducting. These critical current data are in agreement with SQUID measurements of the irreversible magnetization using the Bean model [23] for determination of J_c. Measurements of the upper critical field [14] and the fluctuation conductivity [24] are well understood and indicate that *superconducting coupling* (either by the proximity or the Josephson effect) can be excluded as the origin of the minimum. Consequently, any reference below to *coupling* will relate to *magnetic* coupling between the flux lines in the Pb layers.

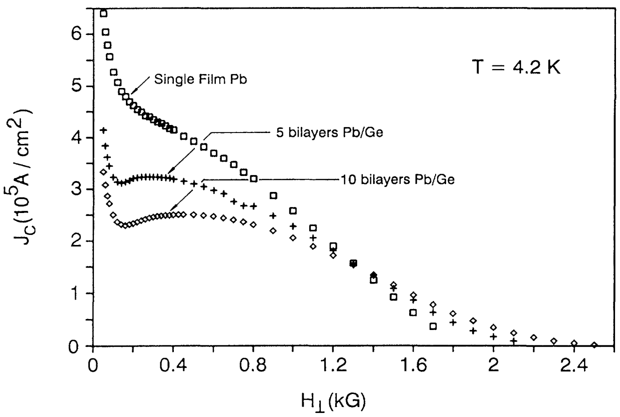

FIG. 1. Critical current density J_c, measured resistively, as a function of perpendicular field for three [Ge(60 Å)/Pb(140 Å)]$_n$ structures with a different number of bilayers [□ ($n=1$, single film), + ($n=5$), and ◇ ($n=10$)].

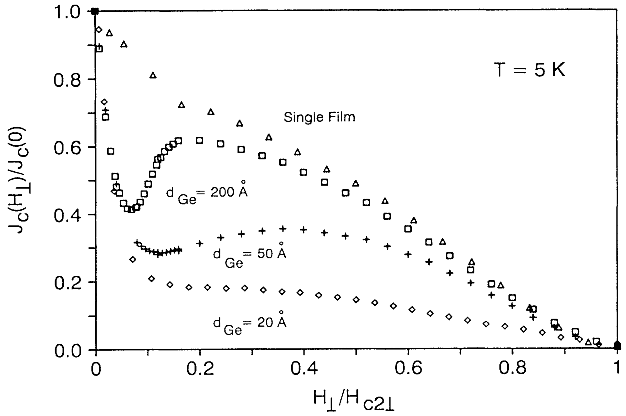

FIG. 2. Normalized critical current density vs $H_\perp/H_{c2\perp}$, determined from magnetization measurements, for three [Ge/Pb]$_{50}$ multilayers with $d_{Pb}=200$ Å, but different d_{Ge} [□ ($d_{Ge}=200$ Å), + ($d_{Ge}=50$ Å), and ◇ ($d_{Ge}=20$ Å)].

The critical current density J_c, measured resistively, of a single Pb layer [i.e., a Ge(60 Å)/Pb(140 Å)/Ge(500 Å) sandwich] decreases monotonously with applied perpendicular field $H_\perp$, as expected naively from standard superconductivity theory (Fig. 1). An interesting nonmonotonous behavior develops in the multilayers. As a multilayer is built up, a minimum in $J_c(H_\perp)$ appears at a field $H_\perp^*$, followed by a broad maximum. J_c joins the critical current density of the single Pb film for fields $H_\perp \gg H_\perp^*$, as shown in Fig. 1 at $T=4.2$ K for [Ge(60 Å)/Pb(140 Å)]$_5$ and [Ge(60 Å)/Pb(140 Å)]$_{10}$ multilayers.

This minimum is quite sensitive to the Ge separator thickness d_{Ge}. Figure 2 shows $J_c(H_\perp)/J_c(0)$, obtained from magnetization measurements, versus $H_\perp/H_{c2\perp}$ at $T=5$ K for three multilayers with $d_{Pb}=200$ Å and $n=50$ but with different d_{Ge}. For comparison, the single film behavior ($d_{Pb}=250$ Å) is also shown. Note that J_c had to be normalized by its zero-field value because of uncertainties in the superconducting volume and the demagnetization factor. $J_c(H_\perp)/J_c(0)$ of the strongly coupled multilayer with $d_{Ge}=20$ Å drops much faster than that of the single Pb film. With increasing d_{Ge}, a nonmonotonous $J_c(H_\perp)$ dependence is observed, similar to that in Fig. 1. A further increase in d_{Ge} eventually leads to the single film behavior. At low fields, $J_c(H_\perp)/J_c(0)$ of the three multilayers coincide. With increasing $H_\perp$, an upturn occurs and J_c joins that of the single film. The crossover point shifts to higher fields when decreasing d_{Ge}. This indicates that matching effects [25,26] can be ruled out, which is also confirmed by the pronounced temperature dependence of the minimum.

Figure 3 shows the low-field $J_c(H_\perp)$ for the [Ge(50 Å)/Pb(140 Å)]$_5$ multilayer at $T=1.4$ and 5.6 K, derived from I-V curves. At low temperatures the minimum disappears with the perpendicular field dependence of J_c reverting to the expected monotonous decrease with field. The inset shows the field dependence of $J_c(H_\perp)$ for a [Ge(50 Å)/Pb(100 Å)]$_{10}$ sample compared to $J_c(H_\perp)$ for a [Ge(50 Å)/Pb$_{1-x}$Bi$_x$(100 Å)]$_{10}$ multilayer with $x=0.15$. It is clear that the addition of Bi eliminates the presence of the minimum as does decreasing the temperature. Adding Bi impurities to Pb increases the pinning, which is proven in this sample by the fact that J_c of the Pb-Bi alloy is larger than J_c of Pb. This experiment therefore clearly illustrates that increasing pinning plays a similar role as decreasing temperature. The immediate conclusion is that thermal fluctuations play an important role in the disappearance of the minimum.

At this stage it may be useful to summarize the major experimental findings in these series of experiments: (1)

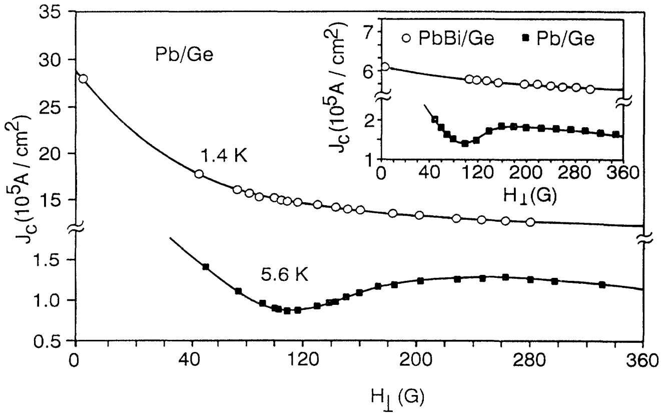

FIG. 3. J_c vs $H_\perp$ for a [Ge(50 Å)/Pb(140 Å)]$_5$ multilayer at $T=1.4$ and 5.6 K, derived from I-V curves. Inset: $J_c(H_\perp)$ for a [Ge(50 Å)/Pb(100 Å)]$_{10}$ multilayer and a [Ge(50 Å)/Pb$_{0.85}$Bi$_{0.15}$ (100 Å)]$_{10}$ multilayer at $T=4.2$ K.

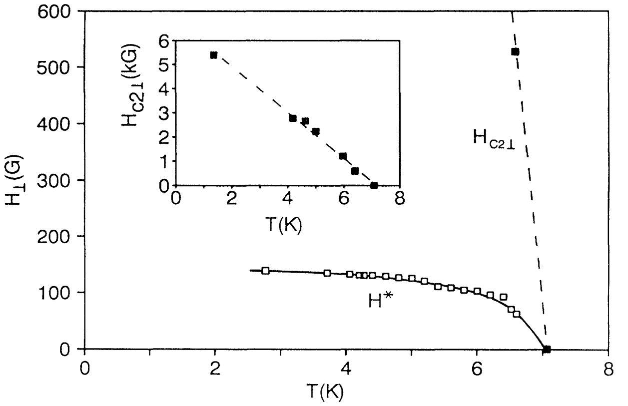

FIG. 4. ($H_\perp$,T) phase diagram of Ge/Pb multilayers. The open squares represent the field $H_\perp^*$ (the solid line through the data points is only a guide to the eye) while the solid squares correspond to the upper critical field $H_{c2\perp}$. Inset: The linear temperature dependence (dashed line) of $H_{c2\perp}$ over the whole temperature range.

The critical current develops a minimum as a function of perpendicular field with increasing number of bilayers; (2) there is an optimum separator thickness for which the minimum is most pronounced; (3) the J_c curve follows the coupled multilayer behavior at low fields and the single Pb film at high fields; and (4) the minimum disappears at low temperature, which also occurs if the pinning density increases. We can summarize all the data in an $H_\perp$-T phase diagram as shown in Fig. 4. The inset shows the temperature dependence of $H_{c2\perp}$, with an expanded view at low fields shown in the main figure. It should be stressed that $H^* \ll H_{c2}$ while it is of the order of $H_{c1} \simeq 20$ G, obtained from magnetization measurements neglecting demagnetization corrections.

To date two classes of theories have been advanced which may explain these results as a consequence of a structural transition in the flux-line lattice. In some general sense the two classes of models can be categorized as "decoupling" and "melting" models. In the decoupling model [16–18], at low fields the vortex-vortex interaction from layer to layer is strong and therefore the flux-line lattice penetrates the whole multilayer. As the perpendicular field is increased, the coupling energy decreases (because of the higher overlap of the vortices) compared to thermal fluctuations and a magnetic decoupling occurs at the field H^*. Once the vortices are decoupled from layer to layer, within each layer the pancake vortices readjust to take advantage of the available pinning centers, thus producing the increase in critical current.

For this model to be applicable, the system has to be in the weak pinning regime, the pinning has to be uncorrelated from layer to layer, and no major changes should occur in the superconducting properties with increasing number of layers. Within this model it is easy to understand the dependence of the minimum as a function of the number of layers, separator thickness, temperature, and pinning. The relative field dependence for a single film and a strongly coupled multilayer as shown in Fig. 2 is more difficult to understand.

Within the melting models in the weak pinning limit, the general dependence of the phase line drawn in Fig. 4 has been predicted [13]. Because of pinning, the vortex liquid which occurs below the phase line is transformed into a vortex solid. The melting occurs because with decreasing field the in-plane vortex-vortex interaction is exponentially weak and so the vortex lattice becomes highly unstable to thermal fluctuations. The melting transition occurs for an anisotropic 3D multilayer when the characteristic length scale γa_v exceeds the separation between the superconducting layers. For the Ge/Pb multilayers discussed here the anisotropy parameter γ is given by the coherence-length ratio $\xi_\perp/\xi_\parallel \simeq 0.1$ [27], whereas the spacing between vortices $a_v \simeq 4500$ Å at H^*, implying that γa_v always exceeds d_{Ge}.

The evolution of the J_c curve is qualitatively understood as follows: As the field is increased ($H \le H_{c1}$) the critical current decreases (down to H_{c1}) because of current-induced depairing. In the extremely low pinning regime, J_c should be zero in the region $H_{c1} \le H \le H^*$ and finite for fields $H > H^*$, going down to zero again at H_{c2}. If the pinning is strong enough, this melting transition should be suppressed, whereas for the pinning regime in between these two limits, $J_c(H_\perp)$ should present a minimum. This is supported by the fact that with increasing pinning the minimum becomes shallower and the minimum moves to higher fields. The main difficulty with this model is that the role played by the layered nature of the material is not clear. Since within this theory the main parameter determining the transition is the perpendicular penetration depth $\lambda_\perp$, all dependences may be attributed to changes in $\lambda_\perp$ although this is somewhat artificial.

Further experiments, including vibrating reed, microwave dissipation, imaging of the flux lattice, specific heat, and further transport measurements, are under way to answer some of the questions raised above.

In summary, we have observed an unusual field and temperature dependence of the critical current of specially engineered artificially layered Ge/Pb semiconducting-superconducting multilayers. The highly reproducible dependences as a function of thickness, temperature, pinning, and field can be qualitatively understood within melting and decoupling models. Very interestingly, the transition is driven by the weakening of the vortex-vortex interaction and not by increasing entropy as observed ordinarily in phase transitions.

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