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Introduction

The study of Artificially Layered Superconductors (ALS) started more than 20 years ago with the search for unusual[1-3] and/or high temperature superconductivity in a variety of metal-semiconducting layers.[4,5] Renewed interest was motivated by the advent of novel preparation techniques that allow control of layer thicknesses close to interatomic distances. In this way layered superconductors can be used as model systems to study a variety of physical phenomena, prepare structures with improved properties and discover novel metastable phases which do not exist in nature. Examples of these studies include: a diversity of dimensional transitions, interaction between superconductivity and magnetism, interaction between superconductivity and electron localization, enhancements of critical fields and critical currents, and the study of incommensurate systems.[6-9]

Recent developments in high temperature ceramic superconductors further increase the importance of studies of Artificially Layered Superconductors. The newly discovered ceramic superconductors are structurally layered and therefore many of their properties will also be determined by this structure. Because of this, particularly in the search for the mechanism of superconductivity, it is important to understand which properties are a consequence of the layered nature of the material and which are due to the presence of some unusual, yet undetermined physical phenomena.

What makes Artificially Layered Superconductors especially attractive for investigation? The main reason rests on the fact that the characteristic lengths which determine the superconducting properties, i.e., the coherence length and penetration depth, are quite long in conventional low temperature superconductors. Existing techniques allow the preparation of layered structures which are perfect at length scales which determine the physical properties. Consequently it is possible to prepare model systems that approach ideal conditions and therefore become an excellent test ground for theoretical models. As in all fields of materials science, unexpected interesting phenomena are also discovered which cannot be predicted *a priori*.

This article will briefly describe preparation and characterization techniques for Artificially Layered Superconductors, the bulk of the article dedicated to describing the superconducting properties including critical temperature, critical current, penetration depth, and critical field.

Preparation and Characterization

With the recent developments in ultrahigh vacuum technology it is possible to prepare highly controlled thin films, and to probe the effects due to a very regular and contamination-free interface. The type of material, growth rates, substrate temperature and contaminant partial pressures are some of the parameters that affect the quality and properties of the multilayers. Currently two major techniques are used to produce multilayered systems — sputtering and thermal evaporation. General reviews and references are available in the literature for each of these techniques.[10-12] Because the details of these techniques are generally known we only present a comparison and show some of the advantages and disadvantages of each.

Sputtering systems have evolved to advanced variants of triode-supported plasma, magnetron, and ion-beam sources which are characterized by high deposition rates (20–100 Å/s). A substantial problem related to sputtering is the interaction between the sample and the plasma, due to secondary electron impact, inert gas ion and atom bombardment as well as vacuum ultraviolet irradiation. These interactions give rise to substrate heating and interface roughness because of intermixing and non-ideal growth modes. An interesting and useful feature of sputtering is the thermalization of atoms which enables control of the energy distribution of impinging atoms as well as easy control of the deposition rate.

More recently high-quality multilayers have also been fabricated by Ultrahigh Vacuum (UHV) Molecular Beam Epitaxy (MBE) techniques using Knudsen cell and electron beam evaporation. The Knudsen cell allows a low, constant evaporation rate (0.1–10 μm/h) enabling accurate control of the composition profile for materials with a melting temperature lower than 1600°C. Electron beam evaporation allows handling of high melting temperature materials, faster growth rates and avoidance of contamination by the crucible, so that a very pure material flux may be obtained. However, a major problem with electron beam evaporation is the control of the deposition rate due to the large temperature gradients in the crucible (up to 2000°C/cm). This requires sophisticated electronic stabilization techniques such as quartz crystal monitors,[13] or Electron Impact Emission Spectroscopy (EIES) with evaporation rate control of about $\pm 5\%$. Better stability ($\sim 1\%$ can be obtained with a quadrupole mass spectrometer that analyzes the number of atoms in the beam.[14]

Properties of Artificially Layered Superconductors are determined by the microscopic arrangement of the constituents, and therefore a detailed knowledge of the structure is needed. Transmission electron microscopy (TEM) and X-ray diffraction (XRD) are two techniques which have been extensively used to characterize the atomic structure and microstructure of Artificially Layered Superconductors.

XRD, a nondestructive characterization tool, provides information in reciprocal space. Due to the loss of phase information, the data cannot be directly converted to real space, and proper structural modeling is needed to explain the spectra. It is well known that a characteristic periodicity d gives rise to diffraction maxima in reciprocal space, with characteristic spacing proportional to $1/d$. Therefore, the low angle x-ray diffraction spectrum of a multilayer contains maxima with a separation proportional to $1/\Lambda$ (Λ is the modulation wavelength) while the high angle region also contains information on the lattice parameters of the constituent materials. The relative peak intensities give information on the chemical and interplanar spacing modulation.

The length over which the x-rays scatter coherently is limited by imperfections in the growth process, substrate roughness, fluctuations on the layer thicknesses due to the preparation method, and the types of constituent materials. The diffraction spectrum is very sensitive to variations on the interfacial structure and composition, a property which is extensively used to investigate diffusion in multilayers.[15]

Much work has gone into understanding the details of the structure and composition at the atomic level in crystalline-crystalline, crystalline-amorphous, and amorphous-amorphous multilayers. Several extensive reviews[16] explain the different models and their relationship to the preparation method, growth mode, and nature of the constituents. Fortunately enough, the length scales involved in the physics of Artificially Layered Superconductors are much longer than any structural defects present in reasonably carefully prepared samples. Figure 1 shows a low angle diffraction spectrum from a Pb/Ge (crystalline/amorphous) Artificially Layered Superconductor obtained in a standard Θ-2Θ diffractometer using Cu K_α radiation.[17] A series of equally spaced peaks indicates the presence of a well-defined modulation, and the fact that all even-order diffraction peaks have almost zero intensity indicates that these multilayers have minimum interdiffusion or interfacial roughness.

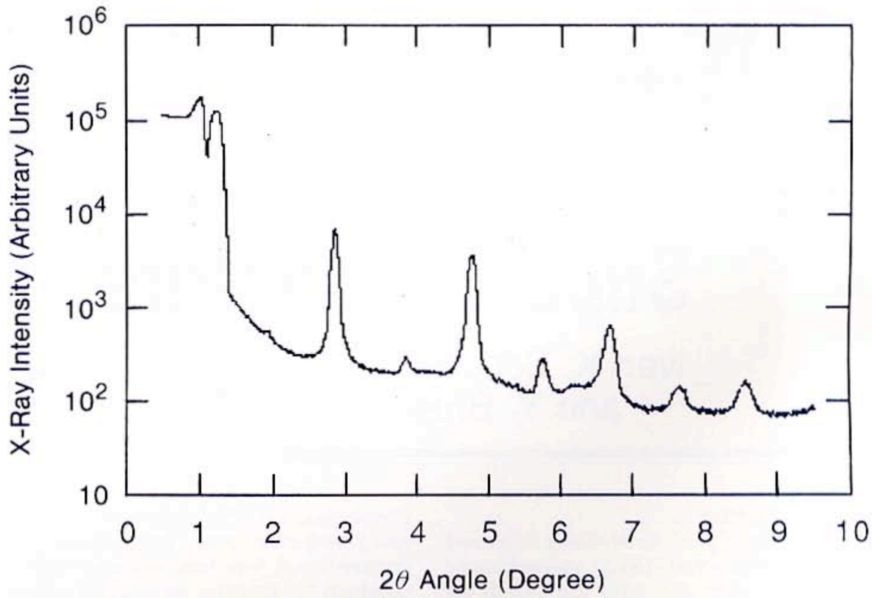

Figure 1. Small angle x-ray diffraction spectrum from a Pb(50 Å)/Ge(50 Å) multilayer (Ref. 17). Note that the even-order peaks are of considerably smaller amplitude than the odd ones.

TEM can provide more local information, although averaging along the electron beam path also occurs and the determination of quantitative information is made difficult by the need of modeling.[18-20] Moreover, TEM requires lengthy preparation of sample cross sections, which may introduce uncontrollable defects. On the other hand, TEM gives a direct real space image of the structure providing feedback information that can be used to improve preparation techniques.

Critical Temperature

The critical temperature of superconducting multilayers has provided[6-10] an interesting field for the study of the interaction between a superconducting and a normal material.[21] Superlattices in which the normal material is either a semiconductor or a metal have both been grown. In the first case the superconducting layers are coupled through the Josephson effect[22] and in the second through the proximity effect.[21]

The case in which the superconducting material is Nb is particularly interesting. Single Nb thin films show a decrease in the critical temperature as the film thickness is reduced. This behavior has been interpreted using two different mechanisms which operate simultaneously. The first one uses the smearing[23] of the Nb *d* electron density of states peak at the Fermi energy due to a finite electron mean free path. In addition to this mechanism, the proximity effect with a damaged or oxidized layer at the surface of the Nb films has been invoked to explain the data.[24]

A detailed study of the dependence of T_c on the modulation length for equal Nb/*metal*(Cu) layer thicknesses[25] was fitted with the de Gennes, Guyon, Werthammer[21] theory of proximity effect. This comparison indicates that the T_c of the Nb layers must vary as a function of layer thickness. This T_c exhibits a less pronounced thickness dependence than the experimental values on single Nb thin films.[24]

A study of Nb/*semiconductor* (Ge) multilayers shows that the T_c depends on both thicknesses.[26] The T_c of Nb, obtained by extrapolating the experimental data to zero Ge layer thickness, is found to be in reasonable agreement with the T_c data extracted from Nb/Cu multilayers.

A comparison of the transition temperatures measured in the three different systems is shown in Figure 2. The fact that the Nb critical temperatures obtained from Nb/Cu multilayers and from Nb/Ge multilayers coincide, may indicate that this is an intrinsic T_c, not dependent on surface damage. Recently Auvil and Ketterson[27] reviewed the Takahashi-Tachiki[28] formulation of the de Gennes-Werthammer[21] theory of obtaining steeper dependences of the multilayer T_c on the modulation length, without the necessity of modifying the T_c of Nb with layer thickness.

Critical Currents

Critical current measurements have been restricted to a few systems including, Pb/Bi,[29] PbBi/Cr,[30] Nb/Ta[31] and NbN/AlN.[32] The critical current density (J_c) of PbBi compositionally modulated alloys, in which the concentration of the Bi component was modulated sinusoidally in the perpendicular direction

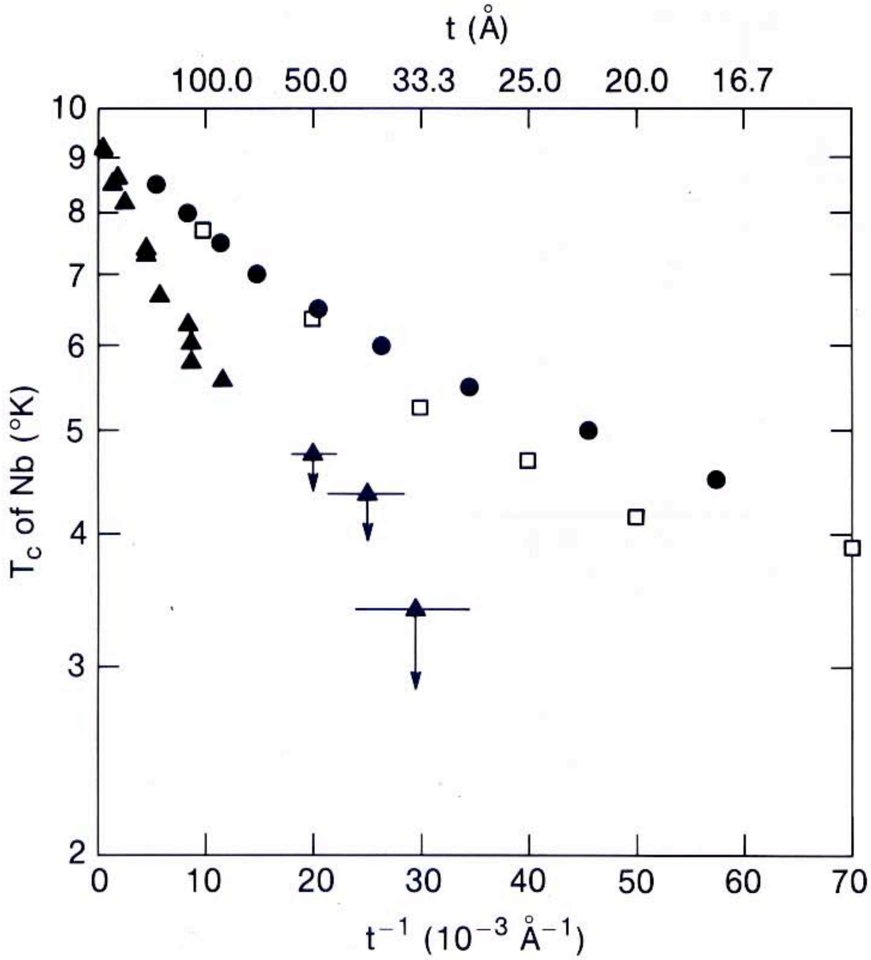

Figure 2. Critical temperature of Nb as a function of Nb thickness, t, obtained (Ref. 24) from single Nb films (triangles), and extracted (Ref. 26) from Nb/Ge multilayers (open squares) and (Ref. 25) Nb/Cu multilayers (closed circles).

to the substrate, shows several peaks as a function of magnetic field (parallel to the equiconcentration layers), indicating an enhanced flux pinning. This is illustrated in Figure 3 for two samples with different periods for the modulation of the Bi concentration. No effect is observed when the field is perpendicular to the layers. Ami and Maki[33] explained this enhancement of the pinning force for a parallel field by a matching of the vortex lattice, with the artificial superstructure giving rise to multiple peaks. When the temperature is higher than a critical value T*, the vortex radius is large compared to the period of the Bi concentration modulation and the typical behavior of a homogeneous alloy with an average Bi concentration is observed. In that case the critical current drops monotonically as the field is increased. The experiments on PbBi/Cr[30] yield information on the flux pinning strength of a single Cr layer with the applied magnetic field parallel to the layers. The flux pinning strength is independent of the Cr layer thickness but drops sharply when the spacing between the Cr layers is smaller than 600 Å because the fluxoid no longer fits between Cr layers.

This effect has been exploited in preparing high critical current density NbN/AlN Artificially Layered Superconductors.[32] This system is particularly attractive since NbN/AlN has a transition temperature which is relatively high for a conventional superconductor and because very high-quality multilayers have been prepared. Large J_c enhancements in fields up to 250 kOe have been observed in Cu stabilized multilayers. At the highest fields, these multilayers had a J_c which was more than 15 times larger than that of specially prepared films of NbN. The enhancement of J_c in these multilayers was found to be due to an enhancement of the pinning forces at the NbN-AlN interface. Thermal stability of these structures was improved by the addition of copper layers interleaved periodically in the multilayer structure. This approach may eventually be extended to high temperature superconductors as well and may result in an increase of J_c.

Finally, high temperature superconductors such as $YBa_2Cu_3O_{7-\delta}$ are excellent examples for the ultimate layered superconductors in which the layering occurs naturally at length scales comparable to the interatomic spacing. In these materials the coherence length is very short and therefore the situation can be described as a multilayer in which the "superconducting" (CuO_2 layers separated by the Y layer) layer thickness is comparable to the perpendicular coherence length, whereas the "normal" (BaO and CuO layers) material layers are somewhat thicker than the perpendicular coherence length. A recent calculation[34] based on superconducting multilayer theory predicts an angular dependence of the critical current in excellent agreement with recent measurements in ceramic superconductors,[35] as shown in Figure 4. These theoretical models do not include any specific features of the ceramic superconductors and therefore could be tested in Artificially Layered Superconductors.

Penetration Depth

The penetration depth λ of a homogeneous superconductor gives direct information on the superconducting order parameter.[36] Its temperature dependence is related to that of the energy gap and Gorkov[37] theoretically found its zero temperature value to be related to the resistivity in the normal state, ρ, and T_c by

$$\lambda(0) = 0.105 \sqrt{\frac{\rho}{T_c}} \qquad (1)$$

where λ is in μm, ρ is in μΩcm, and T_c is in K.

A multilayer can behave as a homogeneous material if the layer thicknesses are less than the coherence length, i.e., if the layers are strongly coupled. However a spatial modulation of the order parameter can subsist[21] and the resulting "homogeneous" material can present anisotropic properties.[22]

The penetration depth has been measured only for the Nb/Cu[38,39] and the V/Ag[40] multilayered systems in the direction perpendicular to the layers, i.e., with the magnetic field applied parallel to them.

Figure 5 shows λ(0) as a function of modulation length Λ for the Nb/Cu system as obtained[38] from magnetic flux expulsion measurements and calculated[41] from Equation 1. The agreement between both sets of data confirms that strongly coupled multilayers behave as homogeneous superconductors. Moreover, in the same experiments the temperature dependence of λ was found to be very well described by the BCS theory in the dirty local limit.

The determination[39] of λ(0) in Nb/Cu multilayers from the field dependence of the Josephson pair tunneling to multilayers agrees with these results for Λ>50 Å. However for Λ<50 Å a decrease in λ(0) is observed instead of the monotonic increase for decreasing Λ predicted by Equation 1. This is presumably related to changes in the multilayer structure at small thicknesses. Kanoda et al.[40] found good agreement between Gorkov's equation and the experimental value of λ(0) for V/Ag multilayers in the coupled regime. The penetration depth was extracted in this case from the temperature dependence of the ac susceptibility. In general, it can be concluded from all these experiments that the dependence of the penetration depth on layer thickness is well described by the Gorkov theory in the coupled regime.

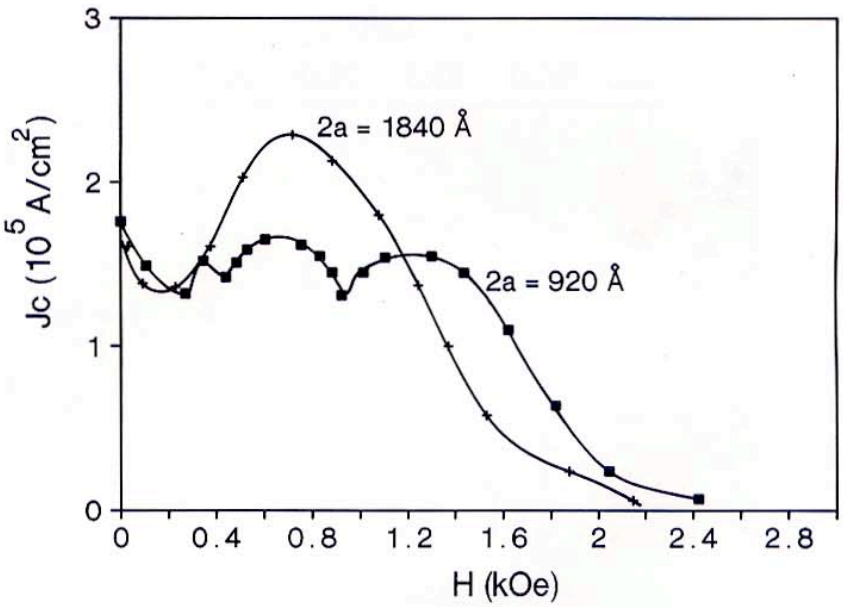

Figure 3. Critical current density vs. parallel field for Pb-Bi compositionally modulated alloys (Ref. 29).

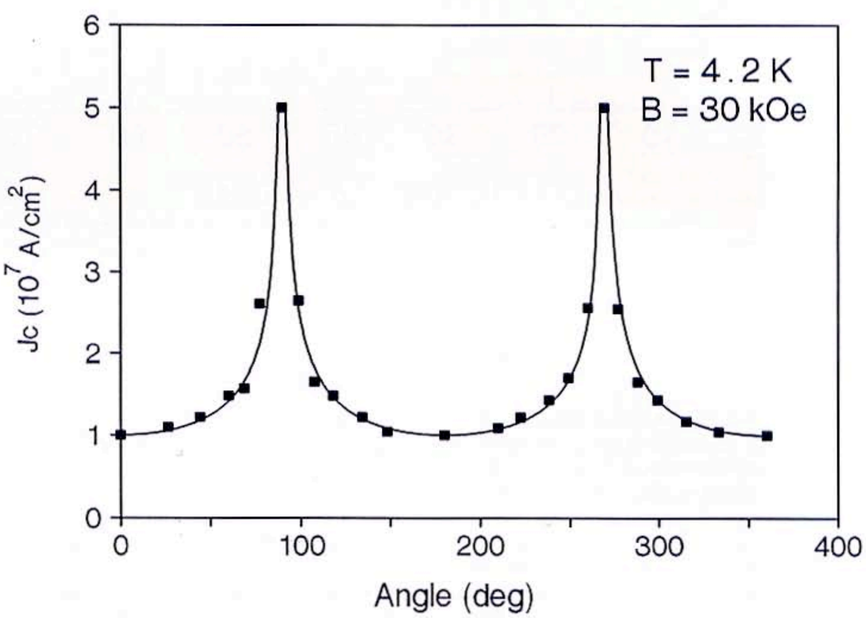

Figure 4. Critical current density anisotropy according to theory (Ref. 34) (solid line), and for high T_c films (Ref. 35) (squares).

Critical Fields

Perhaps the most remarkable properties that have been observed in Artificially Layered Superconductors are related to the superconducting vortices and their structure. At high fields the temperature dependence of the critical field parallel to the layers $H_{c2\parallel}$ shows dimensional crossover, i.e., it changes from the bulk 3D to the 2D dependence as a function of many parameters. In superconducting-ferromagnetic multilayers the $H_{c2\parallel}$ close to T_c is smaller than the perpendicular critical field $H_{c2\perp}$ while at lower temperatures the anisotropy of H_{c2} reverts to the normal one, i.e., $H_{c2\parallel}>H_{c2\perp}$. This anomalous behavior of H_{c2} is quite sensitive to the strength and nature of the coupling between the superconducting layers, i.e., to the interplay between the size of the vortices (superconducting coherence length ξ) and the properties of the layer separating the superconducting thin films. In the very low field region an extreme sensitivity is observed in the behavior of the magnetization of Artificially Lay-

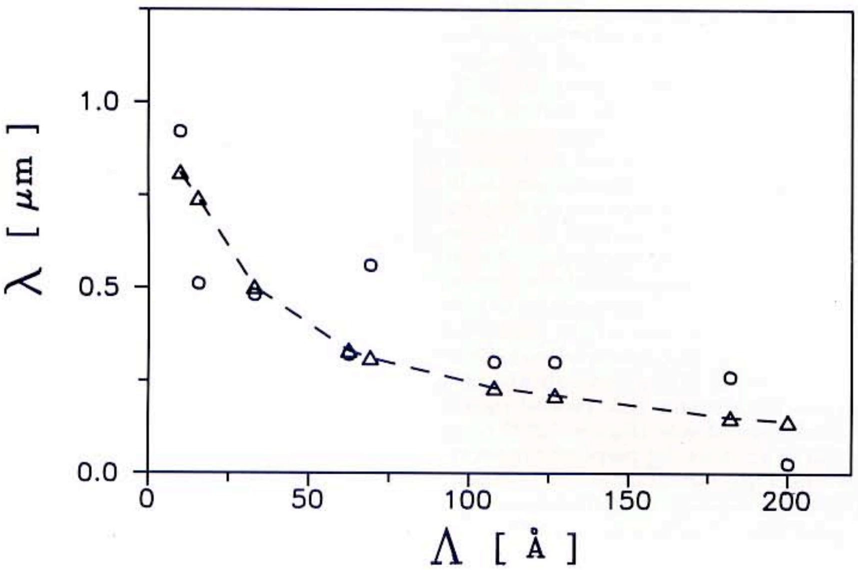

Figure 5. Zero temperature penetration depth λ(0) as a function of multilayer modulation length Λ for Nb/Cu multilayers. Experimental data (Ref. 38) (circles); Gorkov's theory (triangles), Equation 1 (Ref. 37). The dashed line joining the triangles is a guide to the eye.

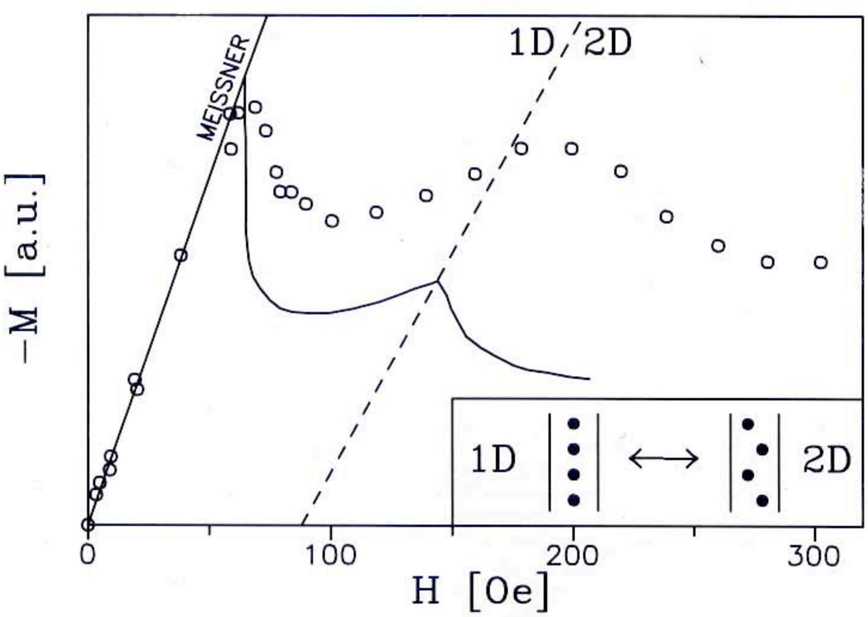

Figure 6. Magnetization as a function of applied magnetic field for a Nb(16.5 Å)/Cu(16.5 Å) multilayer (Ref. 42) with T_c=2.61 K at T=1.7 K: experimental data (circles), theoretical thermodynamic equilibrium magnetization (solid line), theoretical phase boundary between the 1D and the 2D vortex arrays (dashed line). The inset shows schematically the positions of the vortices in the 1D and 2D arrays.

ered Superconductors as a function of parallel magnetic field. A profound study and understanding of the behavior of the vortices, their structure and dependence on the various parameters may considerably impact current activities in natural and artificial layered high T_c superconductors.

One-Dimensional to Two-Dimensional Transition

The superconducting vortex array of Artificially Layered Superconductors has been found to experience a transition[42,43] from a one-dimensional (1D) arrangement to a two-dimensional (2D) one. This transition arises from the competition between the vortex-surface[44] and the vortex-vortex repulsive interaction. The length scale for both interactions is the penetration depth which implies that this phenomenon can only be seen in films with a thickness comparable to λ. Figure 5 shows that λ for the Nb/Cu system is very large,[38] about 1μm, and so it is relatively simple to prepare a thin film with "perfect" surfaces on this length scale.

The dimensional transition is signaled in Nb/Cu multilayers[42] by the atypical behavior of the magnetization shown in Figure 6. At *low fields* the Meissner effect, i.e., the diamagnetic magnetization proportional to the applied field, together with the vortex paramagnetic contribution produce the first maximum. These vortices experience: (a) the repulsive interaction with the surfaces, which keeps them at the center of the film, and (b) the vortex-vortex repulsive interaction, which tends to keep them apart. The resulting stable equilibrium array is a 1D line of equally spaced vortices in the center of the sample (see inset in Figure 6).

At *higher fields* the vortex density and their repulsive energy increases, making it more difficult for new vortices to penetrate the film. As a consequence the vortex density slowly tends to a constant, i.e., a magnetization parallel to the Meissner state. At even *higher fields* the vortex density is enough for the vortex-vortex repulsion energy to overcome the vortex-surface repulsion energy and split the 1D vortex array into a 2D one (see inset in Figure 6). This splitting softens the elastic constants of the array and an enhanced penetration of vortices originates the second maximum in the magnetization. If the field is increased above this point it is theoretically[43] expected that the vortex array experiences successive splittings producing additional maxima in the data.

However, in the measured field range, these transitions are not observed.

Two-Dimensional to Three-Dimensional Transition

One of the most extensively studied effects in Artificially Layered Superconductors has been the transition from two to three dimensions (2D–3D) commonly known as "dimensional crossover" in the critical field versus temperature behavior. This effect has been observed in superconducting-normal multilayers coupled by the proximity or the Josephson effect. The original theory[22] for this effect is based on an anisotropic version of the Ginzburg-Landau theory which includes two different coherence lengths; perpendicular ($\xi_\perp$) and parallel ($\xi_\parallel$) to the layers. The critical fields in this model are given by:

$$H_{c2\parallel}(T)=\frac{\phi_o}{2\pi\xi_\parallel(T)\xi_\perp(T)} \quad (2)$$

$$H_{c2\perp}(T)=\frac{\phi_o}{2\pi\xi^2_\parallel(T)} \quad (3)$$

where ϕ_0 is the flux quantum.

The temperature dependence of the parallel critical field of a single two-dimensional (2D) superconducting film ($d_S < \xi$) is given by

$$H_{c2\parallel}(T) = \frac{\phi_0}{2\pi\xi_\perp(T)d_s/\sqrt{12}} \quad (4)$$

whereas the perpendicular critical field dependence is still given by the Equation 3.

On the other hand, the temperature dependence of the critical field of 3D superconductors is given by:

$$H_{c2}(T) = \frac{\phi_0}{2\pi\xi^2(T)} \quad (5)$$

Since the coherence length diverges as $(T_c\text{-}T)^{-1/2}$ the temperature dependence of a 3D superconductor is linear, whereas a 2D superconductor exhibits a square root behavior.

A set of 2D superconducting films coupled across a normal material will exhibit a 2D–3D transition depending on the ratio of the coherence length to the normal material separator ("dimensional crossover"). For large normal material thickness, the Artificially Layered Superconductors have the physical properties of a set of independent 2D superconducting layers. Therefore, the parallel critical field has a square root dependence in temperature as shown in Figure 7b.[45] For small normal metal separation the 2D superconducting layers couple together to form a 3D superconductor which shows a linear temperature dependence of the critical field (Figure 7a).[45] An interesting situation arises when the separator thickness is comparable to the coherence length. In this case, for high temperatures the long coherence length couples the layers strongly, with the consequent linear temperature dependence of the parallel critical field. As the temperature is lowered, the coherence length decreases and the layers progressively decouple to form a set of 2D superconducting layers, with a square root parallel critical field dependence (Figure 7c).[45] Note that in all cases the perpendicular critical field has a linear temperature dependence since this direction $H_{c2\perp}$ only depends on length scales parallel to the layers (see Equation 3).

Detailed theories[22,28,46] have been developed to explain the temperature and thickness dependence, in addition to the influence of the electron diffusivity and density of states of the constituents. Figure 8 shows a comparison[47] of the parallel critical fields in a variety of systems which have different density of states ratios. The trends regarding the width of the 3D region are in qualitative agreement with the recent theory of Takahashi and Tachiki.[28] As expected the larger the density of states ratio the smaller the 3D region is. A detailed comparison of the critical fields with theory can yield the density of states ratio with an accuracy of ~5% in Nb/Cu Artificially Layered Superconductors. Similar agreement has been found as a function of electron diffusivity[48,49] for the temperature dependence of the critical fields in superconducting/superconducting Artificially Layered Superconductors.

Interaction Effects and Incommensurate Systems

Various other layered structures have been prepared to study interaction effects between competing phenomena such as superconductivity and ferromagnetism and superconductivity and localization. The critical field dependences of V/Ni[50] (*superconducting/ferromagnetic*) Artificially Layered Superconductors are not only determined by dimensional transitions but are also affected by the magnetic pair breaking in the ferromagnetic Ni. Moreover, the Ni layer anisotropy causes an anisotropy in the conduction electron spin polarization and therefore the field induced spin polarization is higher in a

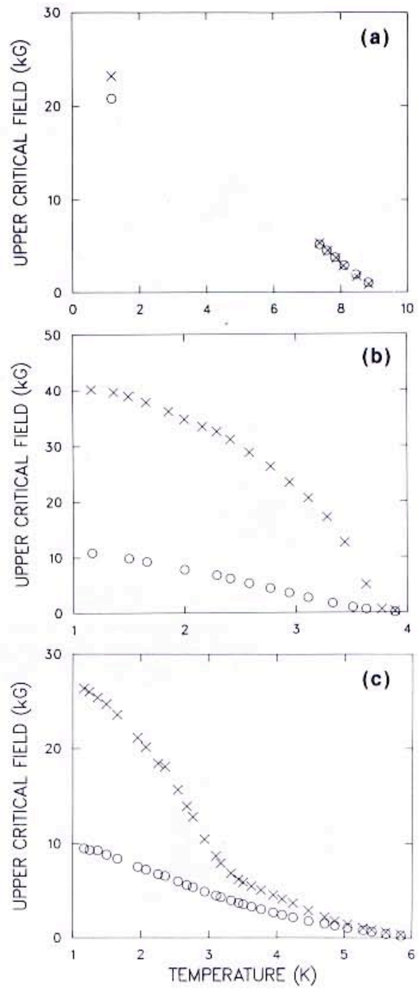

Figure 7. Critical field dependence for Nb/Cu multilayers (Ref. 45) in (a) the strongly coupled 3D regime, (b) the decoupled 2D regime, and (c) the crossover regime. Parallel fields are shown as crosses, perpendicular fields as circles.

parallel than in a perpendicular magnetic field. Consequently, pair breaking due to the spin polarization is greater when the external field is parallel to the layers. The critical field in parallel direction is thus reduced below the perpendicular critical field unlike in any other superconductor, with the exception of the reentrant $ErRh_4B_4$.[51] As the temperature is lowered, the coherence length decreases and the layers decouple. Thus the parallel critical field again reverts to its normal behavior, i.e. $H_{c2\parallel}>H_{c2\perp}$. This type of crossover behavior in the critical

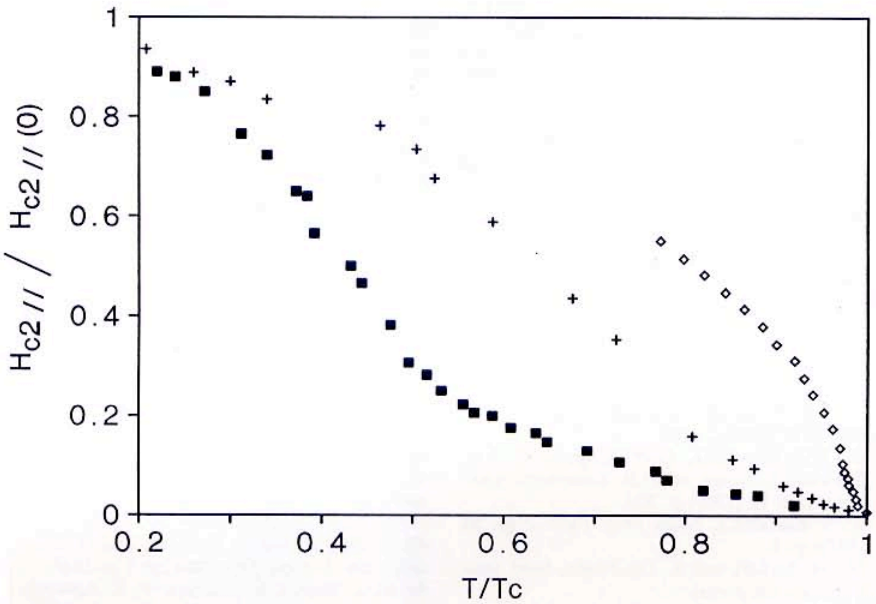

Figure 8. Parallel critical fields in Nb/Ge (open squares) (Ref. 26), Pb/Ge (crosses) (Ref. 47), and Nb/Cu (closed squares) (Ref. 45) Artificially Layered Superconductors showing the influence of the decreasing density of states ratio on the temperature dependence. Ratios of the densities of states are $\frac{N_{Ge}}{N_{Nb}} \leq 0.01$, $\frac{N_{Ge}}{N_{Pb}} \approx 0.05$, $\frac{N_{Cu}}{N_{Nb}} \approx 0.15$.

field has only been observed to date in Artificially Layered Superconductors. An unsolved problem remains the critical field of thin Mo/Ni ALS[52] which in principle at least should exhibit a behavior corresponding to the strongly coupled (3D) regime. However, the critical fields are found to be anisotropic and the temperature dependences are square root-like as if the system would be in the 2D regime. Presently, it is not clear whether these unusual temperature dependences are due to an unknown interaction between superconductivity and localization or magnetism, or due to the behavior of surface superconductivity in these strongly scattering systems.

Nonperiodic-fractal multilayers[53,54] have also been prepared, with varying fractal dimensionality, by growing layers in a sequence given by a prescription such as the Fibonacci series. Consequently, the critical fields of these materials have unusual temperature dependences, between the 2D and 3D limits. A continuous crossover from 2D to 3D is observed as the fractal dimensionality is changed in qualitative agreement with simple scaling arguments based on the theory of Josephson coupled superconductors. However, a quantitative comparison indicates that other factors in addition to the Josephson coupling play a role in the temperature dependence of the critical field and the changes in critical temperature.

Conclusions

Artificially Layered Superconductors are ideal systems for studying thin film, dimensional, proximity, coupling and superlattice effects. They also serve as model systems to check theoretical ideas related to the physics of low dimensional and interfacial materials. The layer thickness, presence of large number of interfaces and the strains induced add a large number of variables which enable fine tuning the material properties and creating novel structures and new systems which do not occur naturally. Until now only a few of these phenomena have been explored in detail so that a quantitative understanding has emerged. Many unexplored problems still exist which have received little or no attention. This includes the study of interaction effects, the changes in the density of states due to the added periodicity, the enhancement of critical currents, the increased pinning in ferromagnetic/superconducting superlattices, the size quantization effects in multilayers containing semimetals, the behavior of nonperiodic multilayers and the studies related to Josephson effects where the periodicity may enhance the emitted radiation. Artificially Layered Superconductors hold promise for use in high critical current tapes, high critical field materials, and as high intensity voltage-controlled Josephson radiators. Finally, the advent of high temperature superconductivity in ceramic oxides allows Artificially Layered Superconductors to be used as a testing ground to distinguish properties and theories which are a consequence of the layered nature and others which are due to more exotic phenomena. Moreover this same technique may help to improve and model the properties of high temperature superconductors, as was done for the conventional ones. Much remains to be done!

Acknowledgments

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