Fractal multilayered superconductors

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Artificially prepared multilayered films with periodic and fractal structure, consisting of amorphous superconducting and normal-metal layers, have been made by magnetron sputtering. The structure has been confirmed directly by transmission electron micrographs of cross sections of the multilayers. The observed upper parallel critical field of the superconducting transition is related to the layering geometry.

It is of general interest to discover how familiar physical properties are modified on fractals, in part because of the numerous examples of fractal structures that exist in nature, and also because of their possible connections to inhomogeneous materials more generally. Fractals also, with their fractional dimension, provide an opportunity to study nature in noninteger dimensions. For these reasons, the literature on the growth and properties of fractals is rapidly growing. At the same time there is currently great interest in artificially structured materials, in which prescribed structures on a near-atomic scale can be produced using advanced vapor deposition and lithographic techniques. Artifically structured periodic superlattices and multilayers, for example, have been studied extensively over the past years.[1] Also, more recently, work has begun on artificially structured quasiperiodic multilayers as a way of understanding aperiodic structures.[2,3] Two-dimensional fractal networks of superconducting material have also been made using advanced lithography,[4] as well as percolative films that have fractal structure.[5] Here, we report the use of recent advances in materials perparation in order to make artificially structured multilayers with a variety of structures, and in particular fractal structures, in order to study physical phenomena in novel geometries.

Fractals are characterized by a self-similar structure, i.e., a structure that repeats on all length scales.[6] Any fractal structure in nature, however, has a finite number of scales over which this structure persists. On the other hand, physical phenomena have their own relevant length scales. It is important therefore to match these physical length scales to the range of the length scales in the model fractal structure.

Here, in particular, we investigate superconducting properties of multilayers consisting of alternating superconducting and normal layers. We have chosen to keep the superconducting layers at a constant thickness and vary the normal-metal thickness within the multilayer. The way in which the normal-metal thickness is varied determines a one-dimensional layering lattice, whose sites are occupied by the superconducting layers. Using computer controlled deposition, the possibilities are very rich. The types of lattices that we have actually made are shown in Fig. 1. They include periodic, "doubly" periodic, random, and fractal. Given these structures, one can then use the temperature-dependent superconducting coherence length to probe the structures. The thicknesses of the layers varied from 10 to 1000 Å, which was determined by consideration of the scales that we could "see" in our measurements as the coherence length was varied by changing the temperature.

Our fractal lattices are all of the same type, created by a cascade procedure similar to that for the triadic Cantor dust.[6] We start with an initiator segment and then cut out a middle portion creating two segments, each being r times the length of the original initiator. This process is then continued on each remaining segment. At each stage of

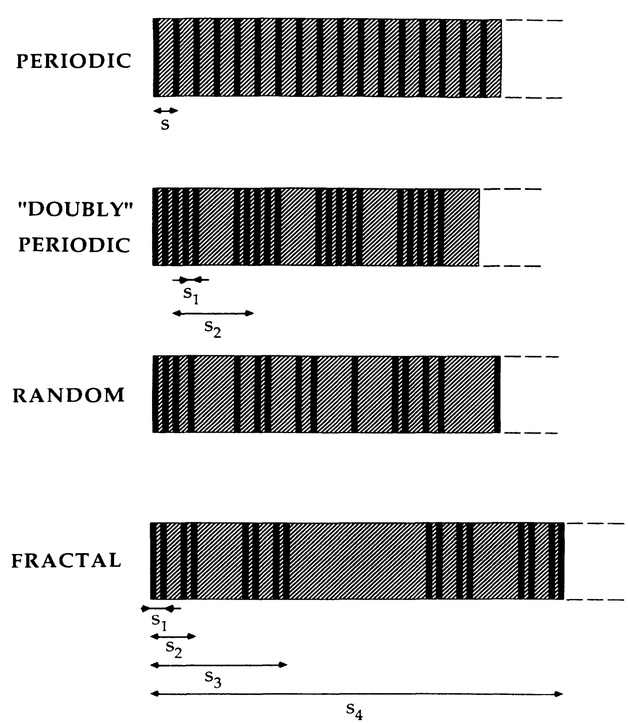

FIG. 1. Superconducting layers in various one-dimensional geometries considered here.

the process the length of the segment is reduced by a factor r ($\frac{1}{3}$ for the triadic set) and a gap is formed of length $1-2r$. By varying r ($0<r<\frac{1}{2}$) we can vary the Hausdorff-Besicovitch (fractal) dimension,[6] $D=\ln 2/\ln(1/r)$, and can then achieve any dimension between 0 and 1. Our layered fractal samples are thus completely characterized by the fractal dimension of the layering D, the number of scales of the fractal structure in the sample, and the thickness of the individual superconducting layers, s_0.

For our system we used amorphous $Mo_{1-x}Ge_x$ of two different compositions, $x \sim 20\%$, with bulk $T_c \sim 7.5$ K, for the superconducting layers and $x \sim 60\%$, with bulk $T_c < 1$ K, for the "normal" layers. These films were magnetron cosputtered onto a-Si_3N_4 substrates, with a thin a-Ge underlayer, in order to grow the films on an appropriate amorphous base, and also a protective overlayer of a-Ge. Substrates were on a continually rotating table in order to achieve uniform coverage and the deposition rates are less than a monolayer per revolution. The layers of the two compositions were put down alternately via computer control of the target shutter. In this way we could make multilayers in any prescribed layering geometry.

Single-layer a-(Mo-Ge) films of various compositions and thicknesses prepared in this fashion have been studied much in the past[7] and have been shown on the basis of transport properties to be homogeneous down to 10 Å. It has also been found that the T_c in these single films is depressed substantially from the bulk value as the thickness of the film is reduced. This depression has been attributed to localization and interaction effects.[7] However, in a multilayer structure it is not clear what the T_c of the superconducting layers should be, as we will discuss later.

A series of samples with periodic and fractal layering was made. We confirmed the structures by cross-sectional transmission electron microscopy (TEM). The cross sections were prepared by ultramicrotomy[8] (i.e., cutting the sample with a microtome blade) and by the conventional mechanical polishing and ion milling technique.[9] Figure 2 shows the micrographs of cross sections of a periodically layered sample and a fractal one, done by each technique.

The cross-sectioned periodic sample has 180-Å layers. The darker regions are Mo-rich (superconducting) layers, while the lighter ones are Ge-rich (normal). Both layers are amorphous, as expected from the deposition conditions, and the contrast between the layers is mass contrast. A small-angle scattering (SAS) pattern from the TEM showed sharp spots due to the bilayer periodicity. The fractal sample shown in Fig. 2 has a layering dimension of 0.88. The self-similar structure can be seen on five scales in this sample. The Mo-rich layers here are 100 Å thick and the Ge-rich layers vary in thickness between 9 and 1000 Å. Note that the interfaces in both samples are sharp and smooth. The deposition rate appears well controlled and the thickness is very uniform in lateral extent. Even the thinnest Ge-rich layer (only 9 Å) can be seen quite clearly through the few-hundred-angstrom-thick section.

We note that the sections prepared by ultramicrotomy offer large viewing areas that extend from top to bottom of the film (in both cases here 4000–5000 Å) and by viewing several sections cut at one time, one can examine the film structure even more extensively to determine its homogeneity. This technique does, however, introduce some artifacts such as the obvious compression "wrinkles" perpendicular to the layers; see Ref. 7 for more discussion of this technique.

X-ray diffraction was also performed on selected sam-

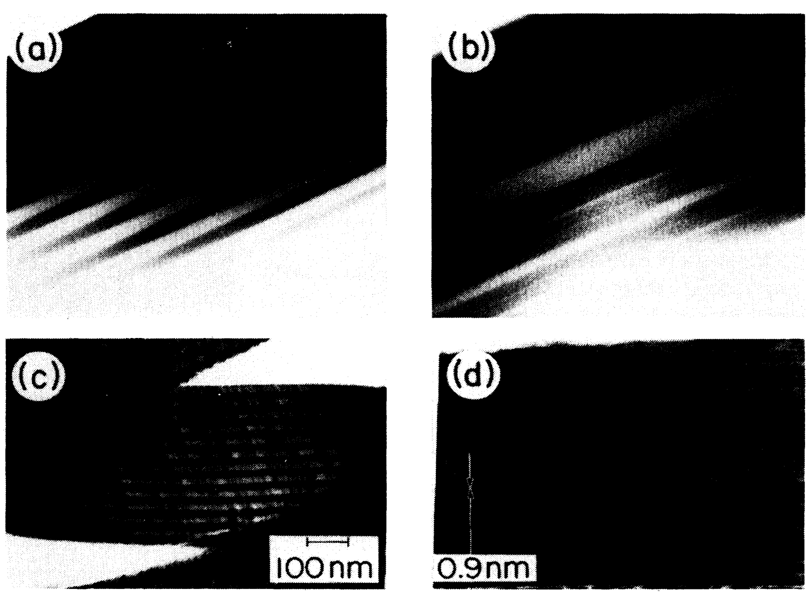

FIG. 2. TEM micrographs of cross sections of a periodic (on the left) and a fractal multilayer (on the right), each prepared in two ways. The top micrographs (a) and (b), are of sections prepared by the mechanical polishing and ion milling technique and were done by Robert Byers at Stanford. The bottom sections (c) and (d) were prepared by ultramicrotomy performed by Ann Marshall at the Stanford Center for Material Research. The interlayer spacing for the periodic multilayer in (a) and (c) is 360 Å. The smallest Ge-rich layers of the fractal multilayer in (b) and (d) are 9 Å thick.

ples. Since these samples are made up of amorphous layers, only the low-angle data should show features due to the layering. Figure 3 shows the low-angle x-ray diffraction data of representative periodic and fractal samples. The periodic sample exhibits low-angle reflections corresponding to the layer repeat distance s; $s=40$ Å in this sample. From the width of the low-angle peaks one can estimate the rms deviation, $ds/s \sim d(2\theta)/2\theta$, to be less than 5%. However, this system is not ideal for x-ray study because of the relatively small difference of the scattering power between the two layers, the intensity of the peaks being proportional to the square of this difference.

A similar x-ray diffraction scan for a triadic-Cantor-set fractal sample shows a hierarchy of peaks. We correlate the largest peaks to the smallest spacing of the layers in this fractal (here 40 Å), the next order of peaks with the next scale, which is three times the previous, and so on. These data agree qualitatively with what one would expect for a Cantor set[10] where there is a hierarchy of weak sidebands, assuming that we have not resolved the higher orders. The slight asymmetry of the sidebands is believed to be due to imperfect adjustment of the deposition rates to produce an ideal triadic Cantor set. High-angle x-ray analysis showed no signs of crystalline inclusions in the samples.

The upper parallel critical field H_{c2} for periodic multilayered superconductors has been much studied in the past.[11] One observes two types of behavior, qualitatively, depending on the relative magnitudes of the superconducting coherence length and the interlayer spacing. When the coherence length is large enough that the order parameter averages over the whole system, bulk anisotropic behavior is observed. This is characterized by linear temperature dependence of H_{c2} near T_c. However, if the coherence length is smaller than the spacing, then the critical field is determined by the single-film two-dimensional (2D) behavior, i.e., a square root temperature dependence. It is well known that the coherence length can be used in this way as a caliper of the structure in the system. Thus, in the case of the periodic multilayer, when the coherence length is on the scale of the variations of the material, one observed experimentally a crossover from 3D to 2D behavior. In the fractal, we would therefore expect to see structure on successive length scales.

Critical-field data were taken on a number of samples with different geometries. Some of the data is shown in Fig. 4. $H_{c2}(T)$ is defined consistently by the midpoint of the superconducting transition. The critical field and the temperature are normalized on this plot to take out any materials dependence and reveal only the structural dependence. In Fig. 4(a) data are shown for a periodic sample in which the coherence length is always larger than the spacing where we see the expected linear behavior. The critical field data for fractals of different dimensions are also shown. D refers to the layering dimension (1 for periodic and 0 for single film). The same data [from Fig. 4(a)] are shown on a log plot in Fig. 5 to reveal any power-law dependence. The data that are shown are not very sensitive to the choice of T_c, which from experimental considerations we believe to be correct to within 2%. One can describe the data over this temperature range by power laws, $(1-t)^{\gamma}$, and assign critical-field exponents γ where γ varies between 1 and $\frac{1}{2}$. We see a systematic decrease of the critical-field exponent with decreasing fractal dimension. The perpendicular critical field, however, shows the usual linear T dependence in all the samples,

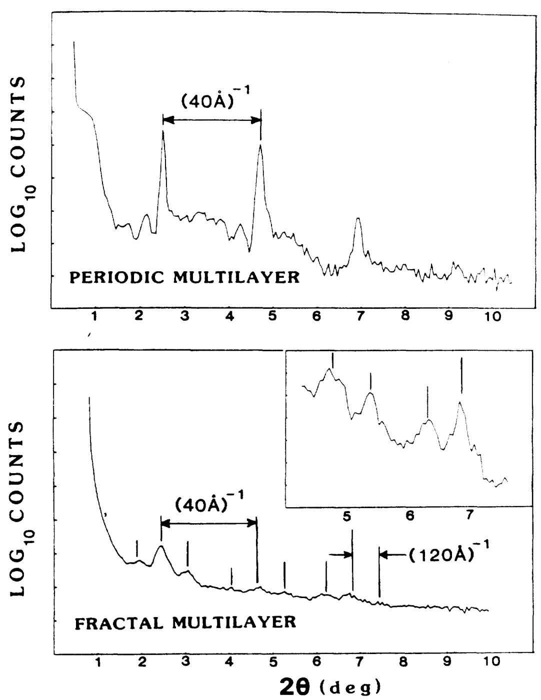

FIG. 3. Low-angle x-ray diffraction scans of periodic and fractal multilayers. The inset on the fractal data is a more careful scan to show the peaks.

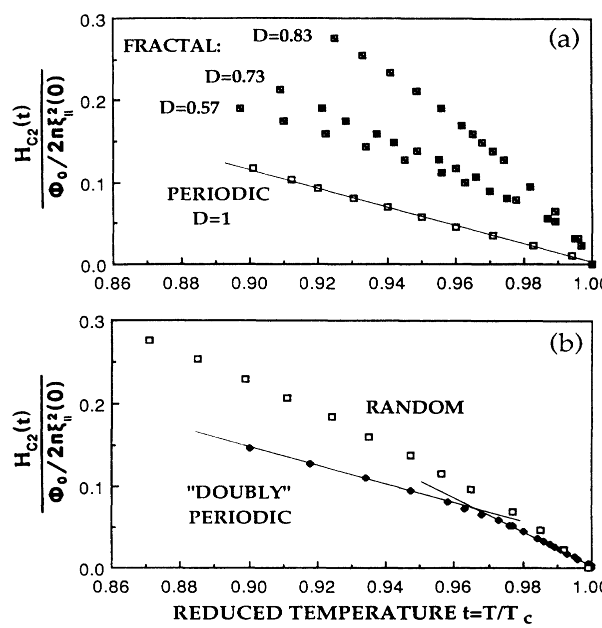

FIG. 4. Upper parallel critical-field H_{c2} data for representative samples of various geometries.

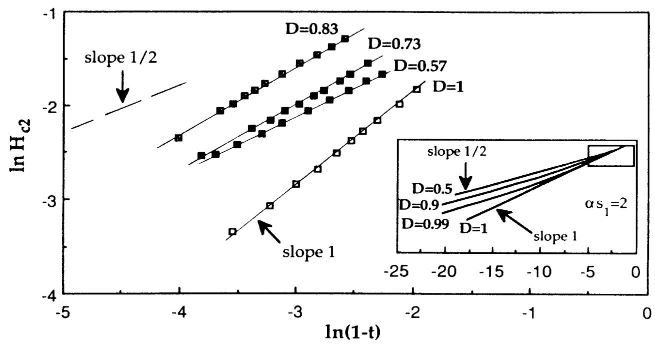

FIG. 5. Log plot of the critical-field data from Fig. 4(a). The inset shows the results of the scaling model calculations. The lines on the data for the fractal samples are a fit using the scaling model.

from which one can extract the parallel coherence lengths. The specific material parameters of the samples shown in Figs. 4 and 5 are given in Table I. The parallel coherence lengths given for each of the samples are derived from data of the same sample set.

In Fig. 4(b) we show the data for the "doubly" periodic and random samples. The doubly periodic sample is made up of six groups of ten closely spaced superconducting layers separated by normal metal spacings of 250 Å. The critical field of the doubly periodic sample shows two linear regions with a crossover corresponding to when the temperature-dependent perpendicular coherence length is roughly 250 Å. The random sample, on the other hand, which is made up of superconducting layers separated by randomly assigned thicknesses of normal metal, exhibits almost linear behavior, which corresponds to the average layering dimension of one in the sample.

From the results presented above it is clear that fractal (and other aperiodic) multilayers can be successfully fabricated, and on length scales that clearly affect the superconducting properties of the multilayer. The question remains, however, whether the observed behavior for the fractal multilayers is universal and therefore dependent only on the fractal dimensionality, or whether other nonuniversal factors are important. At the present time the data are insufficient to answer this question entirely from an experimental point of view. In order to gain some insight into the universality of our results theroretically, we have considered a simple, phenomenological scaling theory of the critical fields for fractal multilayers.

We assume that the layers are Josephson coupled and that the physics of H_{c2} is governed by the change in the zero-temperature coherence length as the temperature-dependent coherence length spans a different number of layers in the fractal. This approach is similar to taking a scale-dependent diffusion coefficient,[12] only here the coherence length is related to the strength of the Josephson coupling. Note that the model must be self-consistent, because as the temperature-dependent coherence length changes the number of coupled layes, this will, in turn, change the zero-temperature coherence length. We neglect possible changes in T_c and only consider the change in the critical field slope as a function of length scale. Any changes in T_c should not alter the results qualitatively.

Consider a fractal structure such as that in Fig. 1 (e.g., triadic Cantor set). We assume that the critical field parallel to the layers has the usual bulk anisotropic dependence

$$H_{c2\parallel}=\frac{\phi_0}{2\pi\xi_\parallel(t)\xi_\perp(t)}\ , \tag{1}$$

where the coherence length perpendicular to the layers depends on temperature explicitly in the usual manner near T_c, i.e., $(1-t)^{-1/2}$, and also implicitly because the effective zero-temperature coherence length depends on the length scale s. Specifically,

$$\xi_\perp(t)\equiv\xi_\perp(t,s)\equiv\xi_\perp(0,s)\frac{1}{\sqrt{1-t}},\ t=\frac{T}{T_c}\ .$$

Thus, we define a series of perpendicular zero-temperature coherence lengths $\xi_{\perp n}$ for each scale of the fractal s_n. The crossover from one scale to the next should occur when the temperature-dependent coherence length is comparable to the next scale of the fractal. Label these crossover temperatures t_n, $1-t_n=[\xi_n(0)/s_n]^2$. Assuming Josephson coupling, the zero-temperature coherence length falls off exponentially with the normal metal spacing:

$$\xi_{\perp,n+1}(0)=\xi_{\perp n}(0)e^{-\alpha(s_{n+1}-2s_n)}\ , \tag{2}$$

where $s_n=2^{(n-1)/D}s_1$ and α are related to the material in question, i.e., α is inversely proportional to the coherence length of the normal material. However, $\xi_{\perp n}(0)$ is not directly related to the coherence length of the supercon-

TABLE I. Description of the samples and experimental values of their superconducting transition temperatures T_c, critical-field exponents γ, and the parallel zero-temperature coherence lengths used in the normalization of critical fields.

Sample No.	Geometry	Dimension	Layer spacing (Å)	T_c (K)	γ(exp)	$\xi_\parallel(0)$[a] (Å)
1	Periodic	$D=1$	$s=60$	4.46	1	60
2	Random	$D=1$	$s=60$	4.17	0.84	60
3	Fractal	$D=0.73$	$s_1=50$	3.72	0.65	58
4	Fractal	$D=0.83$	$s_1=220$	5.61	0.74	58
5	Fractal	$D=0.57$	$s_1=80$	2.80	0.60	58
6	"Doubly" periodic	$D=1$	$s_1=40$, $s_2=650$	6.27	1	48

[a]Parallel zero-temperature coherence lengths are derived from samples from the same set.

ducting material. Physically, $\xi_{\perp 1}$ is the effective perpendicular coherence length of the smallest structural unit (i.e., triad) in the multilayer.

The self-consistency equation is

$$\xi_{\perp n}(t)=s_n$$

at crossover, and it defines implicitly $n(t)$, which we now take to be a continuous function of t. Solving for the relation between n and t, we find

$$1-t_{n+1}=(1-t_1)2^{-2n/D} \times\exp\left[-2\alpha s_1(2^{n/D}-1)\frac{2^{1/D}-2}{2^{1/D}-1}\right], \quad (3)$$

which cannot be solved explicitly, but can be evaluated numerically. One can then compute the critical field dependence on temperature. The general results of such a calculation are shown in the inset of Fig. 5 for some representative material parameters, and specific fits to our data, as discussed below, are shown by the solid lines through the data points. For $D=1$ calculations yield linear H_{c2} behavior and in the limit $D\rightarrow 0$ critical field is square root for all temperatures. For $0<D<1$ and far from T_c, or for small lengths, the linear dependence is recovered (due to the smallest scale built into the model). Near T_c, or for long length scales, the behavior is essentially two dimensional with only a weak logarithmic correction revealing the three-dimensional long-range order. The crossover temperature between these two regimes depends on D, as seen in the figure, and the parameter α.

The physical origin of the limiting behavior as $T\rightarrow T_c$ can be understood simply by noting that using $s_{n+1}-s_1\sim s_{n+1}$ in (2) and (3), it follows from (3) that $2^{n/D}\sim \ln(1-t)^{1/2}$. Solving for H_{c2}, we obtain to first order $H_{c2}\sim(1-t)^{1/2}/-\ln(1-t)$, which shows that the logarithmic factor is a consequence of the exponential fall off of the interlayer Josephson coupling.

Clearly, within this model the coupling in the layered direction is very tenuous. Basically, we find that if the coupling falls off exponentially, the behavior of the system is essentially governed by the dimension of the individual superconducting elements (here it is the topological dimension of the fractal, i.e., 2D), independent of its fractal dimensionality.

This model is consistent with our experimental results, assuming that for the range of data available we are in the crossover region, as shown by the rectangle of the inset of Fig. 5. Quantitative fits to the data are shown by the solid lines through the data, using α and the normalization as free parameters, and s_1 and D as given by deposition conditions. The fits yield values for α of $(50\ \text{Å})^{-1}<\alpha<(100\ \text{Å})^{-1}$, compared with the estimated value $\alpha\sim\xi_n(0)^{-1}\sim(80\ \text{Å})^{-1}$. The fit is quite satisfactory, but should not be regarded as a definitive confirmation of the theory. We point out that if this picture is correct, then we are not observing universal behavior (as a function of fractal dimension, for example) and that the particular critical field exponent that we extract from our measurements is in a crossover regime. However, there clearly is a systematic dependence of these exponents as the fractal dimension is varied.

The sensitivity of H_{c2} to the materials properties on various length scales can be seen directly in the case of the doubly periodic sample shown in Fig. 4(b), which has periodicity on two well-separated length scales. The data very clearly show the crossover. Thus there can be little doubt that the curvature seen in the H_{c2} curves for the fractal samples results from the series of length scales in the structure.

To definitively answer the question of whether the behavior seen is the true long length scale result or not, a more precise theory may be required. Also, there are certain features of the behavior of our multilayers which suggest that other factors in addition to the Josephson coupling are playing a role. As we noted earlier, from the work of Graybeal we know that the transition temperatures of isolated thin films of these amorphous superconductors are greatly reduced below their bulk values. For example, an isolated 30-Å film of the superconducting material (21% Ge) used in the multilayers, has a $T_c=3.4$ K. On the other hand, when these 30-Å layers are incorporated into a multilayer, the T_c can be higher. More specifically, T_c's in the range 3.8 K $<T_c<6.6$ K are observed in periodic multilayers consisting of 30-Å superconducting (21% Ge) layers and normal layers (60% Ge) ranging in thickness from 10 to 90 Å. Taking into consideration the proximity effect, which tends to reduce T_c, it is clear that the T_c of the individual layers, when embedded in the multilayer, must be above the value for an isolated film. Tentative examination of this question suggests that the T_c's of the superconducting layers themselves may be very near the bulk value for the material, and over the whole range of normal layer thicknesses (i.e., for different values of coupling). This striking result clearly needs much closer examination before a definitive understanding of our multilayers will be possible.[13] However, there is no evidence that these T_c effects are the controlling factor in the observed critical field curves.

In summary, we have prepared multilayered films with superconducting layers on a fractal lattice. We observe new behavior of the upper critical field that is directly related to the fractal structure of the layering. A continuous crossover from 2D to 3D behavior is observed as the fractal dimension is changed. Using a simple scaling approach one can describe this behavior qualitatively. The model predicts a universal behavior that is almost 2D-like and if the model is essentially correct and applicable to our samples, it follows also that experimentally we are observing only a transition into that regime.

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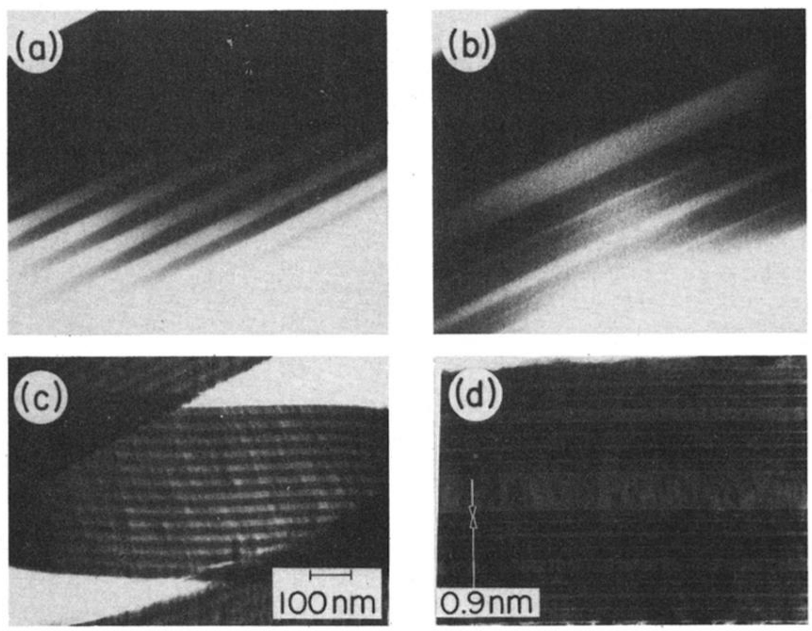

FIG. 2. TEM micrographs of cross sections of a periodic (on the left) and a fractal multilayer (on the right), each prepared in two ways. The top micrographs (a) and (b), are of sections prepared by the mechanical polishing and ion milling technique and were done by Robert Byers at Stanford. The bottom sections (c) and (d) were prepared by ultramicrotomy performed by Ann Marshall at the Stanford Center for Material Research. The interlayer spacing for the periodic multilayer in (a) and (c) is 360 Å. The smallest Ge-rich layers of the fractal multilayer in (b) and (d) are 9 Å thick.