Helvetica Physica Acta, Vol. **61** (1988) 488–495
0018–0238/88/040488–08$1.50 + 0.20/0

Using metallic multilayers to investigate basic physical problems

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(8. II. 1988)

In honor of Martin Peter's 60th birthday.

Abstract. We consider various questions in condensed matter physics that can be fruitfully studied using metallic multilayers as model systems.

1. Introduction

As the technical aspects of vapor deposition systems become more sophisticated, it has become possible to explore a variety of physical problems using artificially structured materials. Epitaxially grown thin films are increasingly being produced and studied and there is no doubt that in the future such materials will gain in importance both for basic studies and for applications. Artificial superlattices (or multilayers-we use the two words interchangeably here) produced by the alternating deposition of ultrathin layers of two elements or compounds is a very attractive way to produce new materials. These have the great advantage over naturally occurring layered structures in that the material parameters may be precisely controlled and varied. It is therefore possible to use them as model systems to investigate various physical phenomena and to test new ideas.

Examples of such studies are well known from semiconductors, but in recent years there has also been a number of investigation on metallic superlattices [1]. Superconductivity is a phenomena that can be investigated in such systems since the modulation wavelength can easily be made of the order of the characteristic lengths of the superconductor. At this point it is interesting to note that the new high temperature superconductors are layered materials and it is not inconceivable that thin film techniques can in the future be used to build up new such materials layer by layer.

The aim of this paper is to investigate some of the issues under discussion in this field, using results from the Geneva group to illustrate some of the points. The multilayers presented in this work were made either by magnetron sputtering or by electron beam evaporation. The details of the preparation are described elsewhere [2].

2. Strain and interdiffusion

A prerequisite for epitaxial growth is close lattice matching between the constituent materials. However it is also of interest to be able to grow superlattices of materials which have a certain difference in the lattice parameters. In these cases a perfect superlattice must be strained, and one question of importance is under what conditions such a perfect strained superlattice can grow and when the lattice will relax by means of misfit dislocations. It has been demonstrated by Philofsky and Hilliard [3] that the transition between the two regimes will take place at a certain modulation wavelength Λ_c below which the lattice is coherent.

This problem has been studied in our group by means of a model developed by Ariosa et al. [4] and experimentally on Mo/V [5, 6] and Nb/Mo [7] multilayers. The model considers the energy density of a partially strained multilayer

$$u = \frac{E}{1-v}\epsilon_\perp(z)^2 + \tilde{G}[\nabla_z d_\perp(z)]^2 + \frac{4u_0}{\delta\Lambda}$$

where the two first terms are the elastic compressional and shear energies of a coherent, finite size area of the superlattice. E and v are Young's modulus and the Poisson ratio, $\epsilon_\perp$ the inplane strain, G a shear-like modulus and $d_\perp(z)$ is the in-plane lattice spacing. The last term represents the energy of the misfit dislocations which limit the size of the area. Here u_0 is the energy per unit length of a misfit dislocation, δ is the average distance between lines of misfit dislocations and Λ is the modulation wavelength. Assuming a given sinusoidal (for simplicity) compositional profile in the z (growth) direction, one obtains upon minimizing the energy with respect to the strain and the number of dislocations, a critical wavelength Λ_c given by:

$$\Lambda_c = 1.766 L_0 \text{ where } L_0 = (u_0/G\epsilon_0^2)^{1/2}, \qquad G = E/2(1+v),\ \epsilon_0 \text{ is the differential strain and we have set } v = \tfrac{1}{3}.$$

For Mo/V and Nb/Mo superlattices, where the mismatch in both cases is about 4%, we obtain $\Lambda_c \sim 70$ Å. And indeed we see, by an analysis of the x-ray diffraction data, a transition from a highly strained state to a state with no strain at a Λ_c of 70–80 Å. In Fig. 1a we show the average strain in a multilayer period as a function of Λ for the Mo/V system [4]. This transition is also seen in the transport and superconducting properties as an anomalous peak in the residual resistance ratio vs Λ and as an anomalous decrease [5] in T_c below Λ_c shown in Fig. 1b. Note the shift in the curve $T_c(\Lambda)$ between the two growth orientations (110) and (001). Since $u_0^2 \sim b^2$ (b is the Burger's vector) we expect Λ_c to be proportional to b. Between the two orientations we expect a shift of $\sqrt{2}$ and

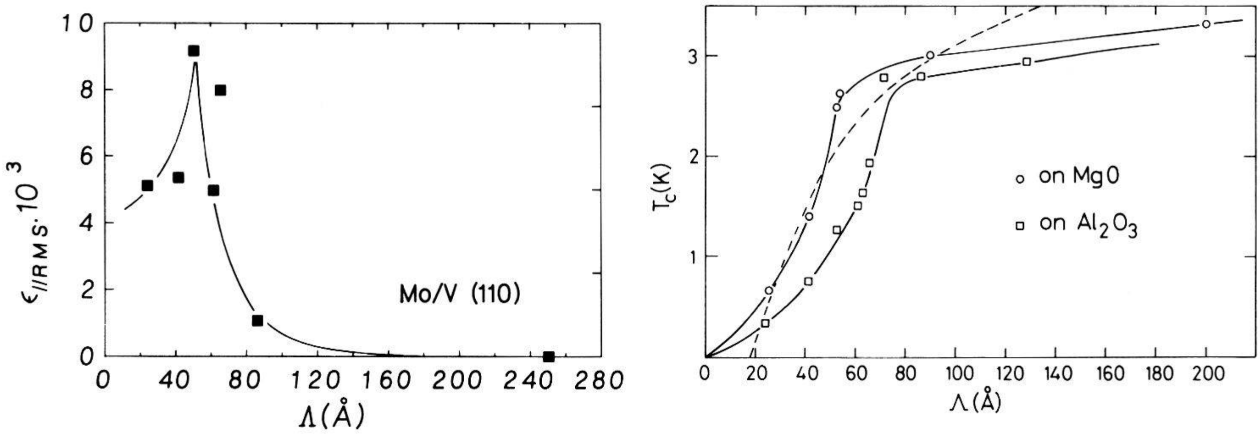

Figure 1
a) The calculated average strain in a multilayer period vs Λ for (110) oriented Mo/V superlattices. b) T_c vs Λ for (110) and (001) oriented Mo/V superlattices. The dashed line is a proximity effect calculation discussed in Ref. 6.

indeed by rescaling the Λ axis by $\sqrt{2}$ for the (001) direction the two curves coincide [5]. In other words the strain behavior depends on the number of atomic planes in the modulation.

3. Upper critical fields

Superlattices contain an obviously built in anisotropy and this can be studied with the help of the upper critical field. If the individual layers are very thin (one or a few atomic layers), then this anisotropy should reflect itself in an anisotropic Fermi surface and electron-phonon coupling. This in turn may lead to anomalous temperature dependence of the upper critical field [8].

In the so far experimentally more accessible region of relatively thick individual layers, the anisotropy is better described by proximity coupling between the layers. Here also, a very non-classical temperature dependence is found and new phenomena is observed. Two such phenomean are illustrated in Fig. 2 where we show the parallel upper critical fields $H_{c2\parallel}$ of Nb/NbTi multilayers. These are made up of two materials having the same critical temperature T_c, but very different diffusion constants. In the 300 Å sample we see a now classical behaviour of dimensional crossover [9]; close to T_c the coherence length becomes very large and the superlattice behaves as a 3-D material with $H_{c2} \propto (T_c - T)$. At lower temperature when the coherence length becomes smaller than the thickness of the individual layers, the clean layers will decouple and behave as a 2-dimensional system, where $H_{c2} \propto (T_c - T)^{1/2}$.

A second and new effect can also be seen in Fig. 2. Takahashi and Tachiki [10] predicted that in a system as the one considered here a phase transition should take place if the layers are thick enough. The basic idea is that the superconducting order parameter can nucleate either in the clean superconductor (Nb) or in the dirty one (NbTi). At relatively high temperatures it is favourable for the system to let the order parameter nucleate in the clean layers which leads

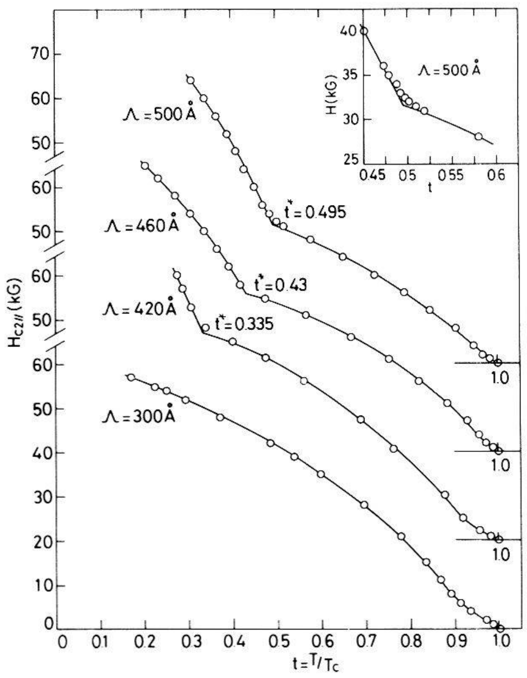

Figure 2
The parallel upper critical field vs reduced temperature t ($= T/T_c$) for $Nb/Nb_{0.6}Ti_{0.4}$ superlattices. The three upper curves show the Takahashi-Tachiki effect while the 300 Å displays only the classical dimensional cross-over. The solid lines guide the eye.

to the decoupled two dimensional behaviour described above. If the order parameter would nucleate in the dirty layers these would be very effectively coupled through the clean layers and this would lead to a lower critical field. However if the layers are thick enough then at lower temperatures the dirty layers will not be much affected by the clean layers and will behave as thick bulk-like films having a high critical field. Takahashi and Tachiki predicted a critical temperature T^* where the order parameter would switch from the clean to the dirty layers leading to a sharp break in the temperature dependence of $H_{c2\|}$. This behaviour is clearly displayed by the samples with $\Lambda = 420$ Å, 460 Å, 500 Å, and, as predicted by theory, T^* increases as Λ increases [11].

4. Magnetism and superconductivity

The interplay of magnetism and superconductivity has been at the center of interest in the field of superconductivity for many years, and the discovery of the new high T_c superconductors has brought additional interest to this field. Superlattices offer unique possibilities to study some of the important issues involved, and the thin film techniques are today approaching a level where it appears feasible to produce such structures with the new materials. Among the questions that are of interest here is the question of how layers of the high T_c superconductors couple through a normal metal or through a classical superconductor. If a different mechanism is present, a behaviour very different from that of superlattices with ordinary superconductors, as for instance described in the previous section, may result.

Several investigations on magnetic and superconducting superlattices have already been carried out with classical superconductors [12]. One of the difficulties here is that in order to test the various problems in an optimal manner it is important that the superconducting and the magnetic state have comparable free energies. If not, one of the states will dominate, there will be no competition, and hence no interesting effects to observe. A problem that would be intriguing to investigate in such structures is the Jaccarino–Peter effect [13, 14]. Indeed, thin films can have very high orbital critical fields, a prerequisite to observe this effect, and using an appropriate magnetic substance one should be able to adjust the magnetic properties correspondingly.

In most simple metals and compounds the magnetic ordering temperature is much higher than the typical superconducting critical temperature. In order to adjust the magnetic transition temperature it becomes necessary to dilute the magnetic system. However here one easily runs into the problem of spin glass behaviour. Long range magnetic ordering can however be obtained when introducing 3d-impurities into Pd or Pt because of the long range of the spin polarization [15]. We have started a program to approach the interplay of magnetism and superconductivity along these lines. We have made superlattices of bcc Nb or *V* with fcc Pd or Pt. The matching is sufficiently good between the *V* and Pt that a partly coherent epitaxial structure can be grown [16]. Here we shall focus on the behaviour of the parallel cirtical fields as shown in Fig. 3 for *V*(110)/Pt(111) superlattices. As can be seen there is no sign of a dimensional crossover as the one clearly displayed in Fig. 2 for the 300 Å Nb/NbTi sample. However, there is a change from 3-D behavior at low Λ to 2-D behavior at high Λ. We speculate that this results from pairbreaking effects due to spin fluctuations in the Pt. A finite lifetime of the Cooper pairs in the Pt will lead to a superconducting order parameter that has a finite 'penetration length' into Pt. Thus, if the thickness of the Pt layers are larger than twice this length they will effectively decouple the *V* layers and the latter will then appear as 2-dimensional even close to T_c. However, in the opposite case of thin Pt layers the *V* layers will

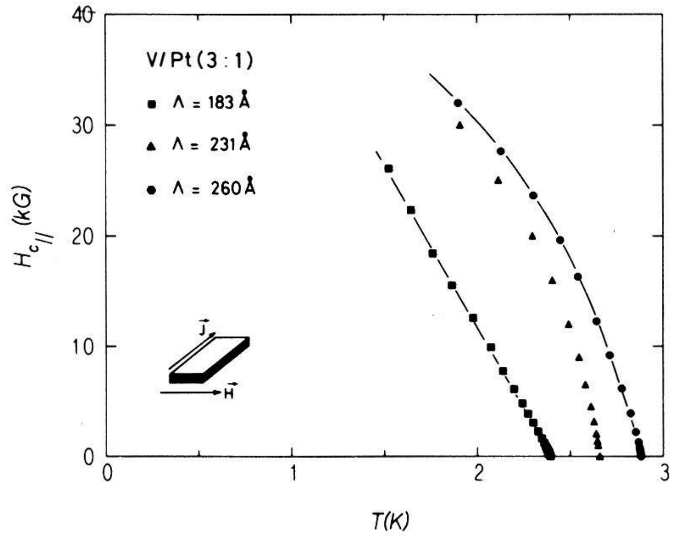

Figure 3
The parallel upper critical field vs wavelength for V/Pt superlattices. The solid lines guide the eye.

couple three-dimensionally at all temperatures. From our measurements this 'penetration length' in Pt is about 50 Å.

To conclude this section let us add a few comments: 1) if our interpretation is correct than we would expect that Pt would behave unlike a normal metal in a typical proximity effect experiment, where T_c is measured as a function of the normal metal thickness, the thickness of the superconductor being kept constant, 2) if V is replaced by a hypothetical superconductor where spin fluctuations are not producing pairbreaking, a dimensional crossover should be observed, and 3) when Fe impurities are introduced into Pt, long range order is obtained and spin fluctuations will be replaced by static polarization effects. Thus a marked change in the behaviour of the critical field would be expected.

5. Quasiperiodicity

Since the discovery by Shechtman et al. [17] of the icosahedral symmetry of quench-condensed Al-Mn, quasicrystals and quasiperiodicity have received a great deal of attention. It has been shown [18] that the diffraction pattern of an ideal 3-D quasicrystal built up from the 1-D quasiperiodic Fibonacci lattice closely corresponds to that of the Al-Mn. Thus multilayers can be used as model systems to study 1-D quasiperiodicity because of its inherent interest and also because it could shed some light on the still unsolved problems of three-dimensional quasicrystals. We have made [19] quasiperiodic (QP) superlattices using the Fibonacci series with Mo and V as the alternating elements in the series. An X-ray diffractogram of a QP multilayer is shown in Fig. 4a and is quite different

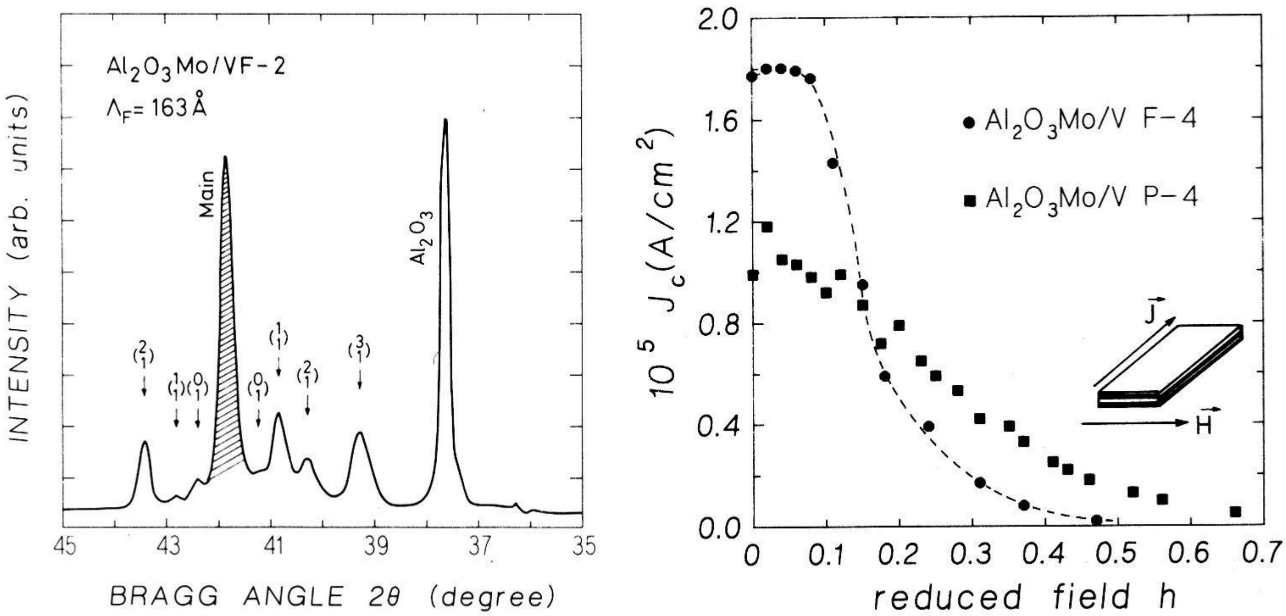

Figure 4
a) X-ray diffractogram for a Fibonacci superlattice. The indexing is explained in the text. b) Critical current density vs applied magnetic field at the same reduced temperature for a Fibonacci superlattice and for a periodic superlattice. The dash line guide the eye.

than a periodic multilayer. Recall that for a periodic superlattice the distance between successive peaks is approximately constant and inversely related to the wavelength Λ_p by $\Lambda_p = \lambda_x/2(\sin\theta_{i+1} - \sin\theta_i)$ where λ_x is the x-ray wavelength. This is no longer true for Fibonacci superlattices. Now the satellites about the main peak can be indexed by $\Delta k = (2\pi/\Lambda_f)n\tau^p$ where Λ_f is the quasiperiodic wavelength, τ the golden mean, n and p are integers, and k is the scattering vector. After having established the difference in the structure of these materials the logical place to look for an effect distinguishably quasiperiodic would be in the interaction of the flux line lattice with the QP layering. (Transport measurements such as resistivity do not yield very much because these are made in the direction parallel to the layers). Fig. 4b shows the critical current density J_c vs applied magnetic field parallel to the layers. We have measured both the QP sample and its periodic counterpart at the same reduced temperature. The two features which stand out are the larger value of J_c for the QP sample at low applied H and the smoothness of the QP curve compared with the P curve. We observe similar behavior for two other pairs of QP and P samples. Even though these results are very preliminary and that as yet there is no theory of critical currents in quasiperiodic media, they suggest that QP could be a useful way to pursue the technologically important subject of critical currents in superconductors.

6. Conclusions

In this paper we have given a few examples that illustrate the variety of basic physical problems that can be investigated using metallic multilayers as model systems. We believe, as the techniques of thin film deposition evolve along with advanced techniques for patterning, that materials produced in this way will be of increasing importance both for basic as well as applied research.

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