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ABSTRACT: After the successful basic solution of the Nb3Ge and Nb3Si problems, optimization strategies and quality criteria were developed for high T_c superconducting (HTCS) samples in analogy. After improvements of the sputtering system, the preparation parameters were tightly controlled, leading to samples with a reproducible linear slope of the specific resistance $\rho(T)$. Further improvements by us and Poppe et al. lead to the preparation of 123 films with non-linear $\rho(T)$ curves above 120K. A generalization to a T^2 behavior of $\rho(T)$ leads to the further generalization that defect free HTCS are essentially two dimensional in nature, with all ensuing consequences. The momentum transport of the moving charge carriers requires, because of momentum conservation, a mechanism to transfer momentum to the lattice (momentum relaxation). The qualitative theory developed for two dimensional (2D) systems shows that the electron-phonon interaction is strongly suppressed and hence that the weaker electron-electron interaction dominates. That theory makes numerous predictions, which so far are confirmed by experiments. In particular, it accounts for the extreme temperature dependence of the mean free path and the Hall effect. It is inferred that the mechanism of HTCS is predominantly due to electron-electron interaction - probably of bipolaron nature, as indicated by experiment. At this point experiment meets just a few theories, that conform with the experimental facts and above theory, all based on the quasi-particle theory of electron-electron interaction. The theory of Newns, Pattnaik and Tsuei should have a T^2 shape of $\rho(T)$ and, because E_F lies near a nearly logarithmic van Hove singularity, accounts for the high T_c. Ruvalds and Virosztek's theory predicts the T^2 shape of $\rho(T)$. Also the marginal Fermi-liquid hypothesis and the Luttinger-liquid theories in 2D make several predictions, in agreement with experiment, except for the imposed linear $\rho(T)$. The task remains to find out, which one of these theories is the correct one.

Qualitätskriterien für Hoch T_c Supraleiter und zur Klärung des Mechanismus der Supraleitung

ABSTRAKT: Nach der erfolgreichen grundsätzlichen Lösung der Nb3Ge und Nb3Si Probleme wurden, in Analogie dazu, Qualitätskriterien für hoch T_c Supraleiter entwickelt. Wesenliche Verbesserungen der Kathodenzerstäubungsanlage gestatteten eine präzise Konstanthaltung der Herstellungsparameter. Dies ergab zunächst Schichten mit reproduzierbar konstanten Steigung der spezifischen Widerstandskurve $\rho(T)$, dann jedoch solche mit deutlich zunehmender Steigung. Poppe et al., in der Verfolgung des Ziels, möglichst perfekte Schichten herzustellen, erhielten dann Schichten, deren nichtlineare $\rho(T)$ Kurven oberhalb 120K mit $\rho(T) = T^k + \rho_o$ angepaßt werden können, mit $k = 1.77$ maximal, wobei ρ_o der Restwiderstand ist. Eine Verallgemeinerung zu einem T^2 Verhalten von $\rho(T)$ führte dann zu der weiteren Verallgemeinerung, daß defektarme hoch T_c Supraleiter im wesenlichen zweidimensionale Schichtstrukturen sind, mit allem was daraus folgt. Infolge des Impulserhaltungssatzes erfordert ein fließender Ladungsträgerstrom Impulsrelaxation, d.h. einen Mechanismus, Impuls auf das Gitter zu übertragen. Die qualitative Theorie, die für Schichtstrukturen entwickelt wurde, ergibt nun, daß dort die Elektron-Phonon Wechselwirkung stark unterdrückt ist, und folglich die schwächere Elektron-Elektron Wechselwirkung dominiert. Diese Theorie ergibt eine Reihe von Vorhersagen, die bisher experimentell bestätigt sind. Insbesondere wird die starke Temperaturabhängigkeit der mittleren freien Weglänge und des Hall-Effekts wiedergegeben. Es wird also gefolgert, daß der Mechanismus der hoch T_c Supraleitung überwiegend auf Elektron-Elektron Wechselwirkung beruht, vermutlich indem sich zwei Elektronen zu einem s-bosonenartigen Bipolaron verpaaren, wie experimentell bestätigt. Dann verbleiben nur wenige Theorien, die alle auf der Quasi-Teilchentheorie basieren. Die Theorie von Newns, Pattnaik und Tsuei sollte ein T^2 Verhalten von $\rho(T)$ haben, und, da die Fermikante nahe einer nahezu logarithmischen van Hove Singularität liegt, wäre auch das hohe T_c, sowie weitere Experimente erklärt. Die Theorie von Ruvalds und Virosztek hat ein T^2 Verhalten von $\rho(T)$. Die marginalen Fermi- sowie die Luttinger-Flüssigkeitshypothesen in zwei Dimensionen ergeben ebenfalls vernünftige Übereinstimmung mit Experimenten, bis auf das postulierte lineare $\rho(T)$. Es verbleibt, zwischen diesen Theorien experimentell zu unterscheiden.

1. Introduction

The title word conceptional is meant to express that before a successful theory can be made, its basic assumptions must be right. Otherwise, even if a single one is wrong, contradictions are likely to arise. So, as shown in this paper, apparently one of the basic assumptions of the marginal Fermi liquid and other theories, namely the linear shape of $\rho(T)$ is not valid. To really understand that conclusion, we must first make a long excursion into the past. If in a hurry, skip to page 8.

It has been pointed out previously [1,2,3], that, if there are 10-15 relevant preparation parameters, at least 10^6 trials are necessary, and reaching the aim of preparing nearly perfectly ordered samples just by trial and error, is very improbable. Optimization strategies are then needed, telling one which property to optimize at each stage of solving the problem. One of the aims of this paper is to discuss, as far as this is presently possible, what are the relevant properties, and with what subsidiary conditions.

For the A15 systems Nb_3X, the importance of preparing homogeneous and nearly perfectly ordered samples, especially thin films, was not realized until interest in those systems nearly stopped, because of the discovery of the HTCS systems. It was recognized that the T_c, the lattice parameter a_o and the at% concentration c correlations in $A_{3+3c}B_{1-c}$ permitted to classify samples according to their ordering. Only a very minor fraction of the samples prepared was homogeneous with optimum values [4,5].

In particular, the specific electrical resistance extrapolated to 0K, ρ_o as a function the B atom concentration c for the A_3B compounds Nb_3Ge, Nb_3Sn and V_3Si curves compiled by Flükiger [6] are then identical within experimental error. They constitute a universal curve that permits to check the homogeneouity and ordering of any sample prepared. At present, this generalization is restricted to A_3B compounds with A elements of the Va and B elements of the IVb group of the periodic system [6,7].

In analogy to above universal curve, another aim of the present paper is to find which properties should have a corresponding universal $\rho(T)$ curve for the $YBa_2Cu_3O_{7-\delta}$ and $YBa_2Cu_4O_8$, and similar systems containing suitable rare earth elements. In analogy to the ρ_o vs c universal curve, the authors understanding of physics leads them to the expectation that reproducibly a linear $\rho(T)$ for above HTCS systems with a minimal slope m is less than 0.6 above the region of the fluctuations and below the temperature, where the oxygen starts to diffuse out ($\approx$650K) should exist, with negative intercept ρ_o. When data for different systems of the same class are plotted as function of the reduced temperature $T_R=T/\Theta_D$, a universal $\rho(T_R)$ should exist, as for monovalent metals [8]. This scheme should hold for conduction in three dimensions. It should also be applicable for systems where conduction takes place in two dimensions, but there are sufficient defects to provide for conduction between planes.

Some more work has to be done to clarify how to handle small changes in the Debye temperature Θ_D. As for Flükiger's universal curve [6] for A_3B compounds, the curves should be nearly identical, when suitable rare earth elements are substituted for Y, as these systems should have a similar Θ_D.

While parts of the chain of argumentation presented above are unchecked so far by experiment, every step is based on arguments of how nature behaves in closely related systems and on the authors belief that general trends observed in other physical systems should also operate in the HTCS field. After all there is much Cu in the 123 and 124 systems. Even if details prove on checking to behave otherwise, the proposed checks provide a rational procedure to advance the aim to prepare optimally ordered, reproducible HTCS samples. This is the formulation of the goal. It should speed up the solution of the HTCS

problem. This appears to us to be superior to the aimless trials documented in many publications. Even if the pursual of the optimization strategies [2,3] will not end with $\rho(T)$ curves, we will then be able to formulate the next step.

Preliminary papers on homogeneous Nb_3Ge and Nb_3Si samples have been previously published [4,5,9]. That work and a brief description of the improvements in the experimental equipment and the optimization strategies persued have been also published [3] and are available as posters in small format. Certain parts of the information in this paper have been presented at the APS 1990 [2]. Parts of the present paper have been presented at the NATO ASI on Applied Superconductivity [10].

2. The Experimental Situation and Discussion

Before starting the HR series of experiments, substantial improvements of the sputtering system were carried out to keep the preparation parameters constant. Regulated power supplies were installed. A new design of the heating element was employed: In order to reduce the temperature difference between substrate and resistive heating element, a flattened stainless steel cylinder with only two small openings was used. The underlying idea is to approach the uniform radiation field in the inside of a black cavity as closely as possibly. The substrate is mounted on a Ni-block, with the a hole for the thermocouple. There are only two openings: One on top for the plasma to reach the substrate and one on the side for the removal of the Ni-block. An alternate solution is a vertical heated cylinder with a open top. Such designs reduce the problem of the film temperature changing when the surface turns black, as the film grows, and hence the radiation losses increase. A PID regulation for the substrate temperature to $\pm 1K$ was installed. For the O_2 and Ar needle valves, a second open/close valve was installed in series, to avoid the drifting, when the setting is not changed. Substrate temperature and total pressure were monitored digitally. Remaining pressure drifts were compensating by changing the entrant pressure. A cylindrical target geometry was employed to minimize impact damage by O^- ions. A substrate temperature gradient was used to substantially reduce the number of runs required. This also has the advantage, that the other parameters are identical for that run. The plasma current was held constant at 0.5A. The pressures from run to run were varied around $P_{O_2} = 0.2mbar$ and $P_{Ar} = 0.4mbar$.

The author has observed [1] that when the substrate temperature T_s and other critical preparation parameters like the O_2 partial pressure are tightly controlled, the T_c downset (zero resistance) and even more critically, the ΔT_c transition widths show sharp minima less than 5K wide in T_s.

A temperature gradient of about 20K along the length of the substrate is used to cut down the number of samples required. Photomicrographs show pronounced variations in the observed structure and the amount of foreign phases, indicating an effective temperature "window" of less than 3K, and a stoichiometry requirement of less than 1-2at%. A nearly perfect film should not contain any foreign phases. In fact our film, which had the least amount of foreign phases, showed no optically detectable structure, except small black CuO deposits of less than 0.1area% (diameter<1μm). This observation means that even an excess of less than 0.1 area% CuO segregates out. This indicates that the 123-system is a real line compound. Hence the stoichiometry must be controlled to probably less than 0.1% , in order to get phase pure films.

Figure 1 (top) shows ρ_o, above ρ_{RT} and (bottom) the slope m for $\rho(T) = \rho_o + m*T$ for the data of the last eight of our samples as function of the substrate temperature, with parabolic fits. The given temperature is probably on the low side, as only reproducibility and no absolute values were aimed for.

The position of the minima decreases with falling O_2 partial pressure. Note that ρ_o and m are minimized simultaneously. Their linear correlation has been noted and attributed to percolation, probably associated with weak links [11]. This strong correlation implies a complete breakdown of Matthiessen's rule [12].

In most cases, three points from one sample prepared with a temperature gradient along the length of the sample, are shown. The circular points on the curve show data from three runs, an indication that the temperature at the surface of the substrate was reproduced to about 1K [54]. The 12K increase of T_S correlated with a P_{O_2} increase by a factor of two (top of figure 1) implies that the P_{O_2} must also be controlled to ±5%. Without T_S control, such curves are widened by the uncertainty in T_S at the substrate surface, (not at a thermocouple anywhere nearby). Also the other top right curve indicates a negative intercept of 40μΩcm between the left and the middle points measured. This amounts to a negative intercept of about -10% of the RT value.

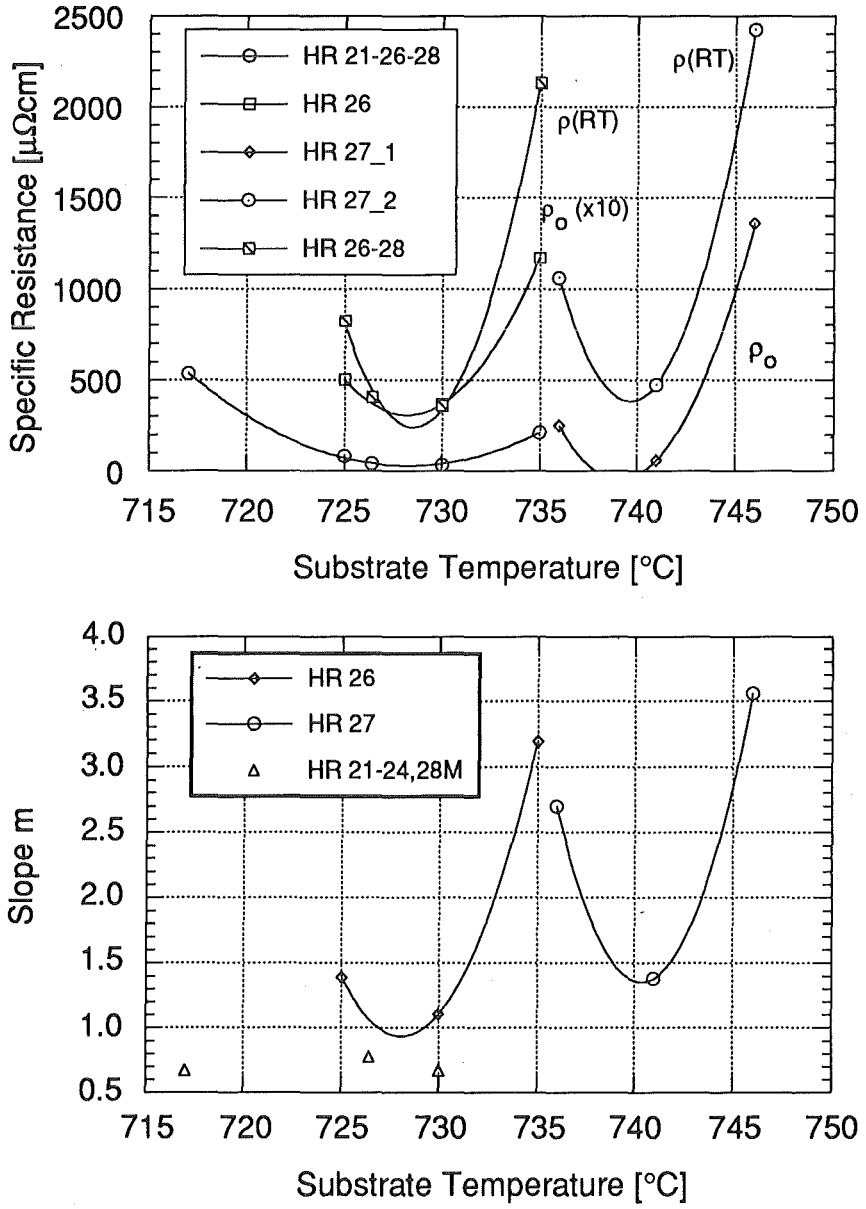

Figure 1. Plot of the slope m (= dρ/dT) (bottom), ρ_o and ρ(RT) (top) versus substrate temperature T_s fitted above 120K. The lines are least squares parabolic fits. Note the excellent reproducibility of the fit through five points from three samples (circles), and the reproducible m values of 0.6 (triangles), which is the value observed for the 124 system. The data points on the right stem from samples, that were prepared at twice the O_2 partial pressure, indicating the necessity to control the O_2 pressure to an accuracy of better than ±5%.

An essential point to make is that the samples have reproducibly the same properties only, if sufficient control of the preparation parameters, like the substrate temperature T_S, the gas pressures P_{O_2} and P_{Ar} and others is achieved. Three samples shown as triangles in the bottom of figure 1 have reproducibly a minimum slope around m = 0.6. Out of the 8 samples HR 21-28 prepared, m was reproduced thrice and the five ρ_o-values from three samples lie on the same curve, from which the conclusion is drawn, that control of the substrate surface temperature within about 1K was achieved [54]. Remember that the emission changes, as the film grows opaque. Whitout above design, $\Delta T_S \approx 20K$.

So while figure 1 indicates that both m and ρ_o of $\rho(T)$ should be minimized together, this may be the case only initially. As m and ρ_o become smaller, the number of defects in the samples will go down. Of course this will be accompanied by a reduction of the number of pinning centers and hence the critical current will decrease. As in the case of the A15 materials [6], it will then become necessary to provide additional pinning centers either by suitable atomic substitution, in analogy to the substitution of a fraction of the Nb atoms in Nb_3Sn by Ta [6], or small, that is < 10nm grains of foreign phase [13], or by irradiation, which certainly for thin films is very interesting, as it provides pinning channels of a controllable diameter of < 5nm, without the sample becoming radioactive [14].

However, as the number of defects falls below a critical value, the atomic ordering of the samples will improve and, as is known for the binary systems CuAu [15] or Nb_3Pt [6] the slope of the $\rho(T)$ curves increases with improved ordering, until it asymptotically reaches reproducibly a unique value. It may well be that the same will happen with the HTCS materials.

A word of caution about the 123 compounds is necessary. It has been argued [16] that, as they have only one pair of Cu planes per unit cell (for details, see ref. 44), thought to be correlated with the observed low charge carrier density [17], a reduced Debye temperature might be effective, as for some poor metals like Bi. However, Θ_D=120K for Bi corresponds to its softness and is quite similar to that of comparable metals. The lowest known Θ_D for metals are 69K (Hg), 89K (Te) and 90K (Pb). These values give a negative intercept of some % of the RT values.

This argument is certainly not applicable for the systems with two or three CuO_2 pair of planes (+ CuO chains) like 124 [17] (with a slope of 0.6 and a negative intercept of -22% of the RT value) or for $TlCa_3BaCu_3O_8$ [18] (with a intercept of -8% of the RT value).

The slope of 0.60 is identical with that of our best 123 samples and the ρ_o = -22% is in excellent agreement with the theoretical value of -23% obtained in Section 3. A slope m = 0.5 has also been observed by Batlogg.

Of course it should be realized that the cuprates are highly anisotropic. Channeling measurements of films on (100) $SrTiO_3$ substrates show that the films are oriented with the c-axis perpendicular to the substrate, but probably with random a-b orientation, due to twinning. At least I am not aware of any work that proves a-b orientation by X-ray measurements for thin films. So the slope of 0.6 is apparently a mean value for a mixed a and b orientation, in series with weak links in between, serving as scattering centers. There is also an a-b anisotropy (due to the chains) by a factor of two, which will cause an increased zig-zag path for randomly a-b oriented grains. This a-b anisotropy might cause an increase in the exponent from the expected T^2-dependence for isotropic 2D films, as observed for an intercalated compound [27].

A highly disordered bottom layer with a thickness of about 10nm increases $\rho(T)$ so far, as indicated by an Ar-peak in the RBS spectra there. This is thought to be due to an initially too high density of nuclei. The largest grains grow fastest, leading to the frequently observed columnar growth. What one should aim for, is truly epitaxial layer by layer growth. To achieve this, the use of tetragonal substrates with an initially reduced growth rate, perhaps at an initially lower substrate temperature, might help.

3. The Theoretical Situation and Discussion

The classical theory of $\rho(T)$ for three dimensional systems is that of Bloch-Grüneisen [19,20]. This explains quite correctly the low temperature behavior as T^n for T less than or equal to about $\Theta_D/5$. The experimentally observed n = 2-4 values are caused by the effective low frequency phonon cut-off, explaining the decrease from 5 in T^5. Above, a linear rise above Θ_D and an intermediate region below the high temperature extrapolated line (see figure 3) follow. For the low temperature region, exponents n varying between 2 and 4.1 have been observed so far. The curves, when linearly fitted above about $\Theta_D/2$ have a negative intercept, which depends only little on n (see figure 3), but strongly on Θ_D. For Au ($\Theta_D \approx 153K$), the negative intercept is -5% of the RT value. For $\Theta_D=420K$ ($YBa_2Cu_3O_7$) and n=3.0 the calculated negative intercept is -23% of the RT value and, physically probably more relevant, -17% of $\rho(\Theta_D)$. A comparison between the experimental $\rho(T)$ versus T curve for an Au film having a thickness of about 300 nm with the generalized least squares Bloch-Grüneisen fit shows no visible deviation .

Generalized ($n \neq 5$) Bloch-Grüneisen function:

$$\rho(T) = 4 \cdot \rho(\Theta_D) \cdot (T/\Theta_D)^n \int_0^{\Theta_D/T} \left[\frac{z^n}{(e^z - 1)(1 - e^{-z})}\right] dz$$

Figure 2 (top) shows the $\rho(T)$ curve of an Au film. The Bloch-Grüneisen fit is so good, that the experimental and fitted curves coincide. Only the difference curve on a scale blown up by a factor of 100 shows some structure (see bottom of figure 2). The remaining deviation is probably caused by noise and instabilities in the automatic T_c measuring system. For this $\rho(T)$ calculation n was numerically fitted as the fractional 4.1, the fit parameter was $\Theta_R=153K=\Theta_D$ and with an arbitrary prefactor. The exponent of the low temperature part of the experimental curve can then be fitted as n = 4.0, rather than the 5.0 value given in textbooks. The maximum deviation is less than 0.2%. This proves that the Bloch-Grüneisen theory still provides an excellent parametrization, even though it has been succeeded by first principles calculations [21,22].

The reduction in n for real systems is caused by a lower effective phonon frequency cut-off below 10-15eV. In the Bloch-Grüneisen theory, only an upper cut-off is used. The higher temperature linearity is due to the 3D isotropy assumption of the electron-phonon scattering [20]. This assumption is only approximately correct. In experimental $\rho(T)$ curves, such nonlinearities are definitely observed - e.g. due to thermal expansion [8].

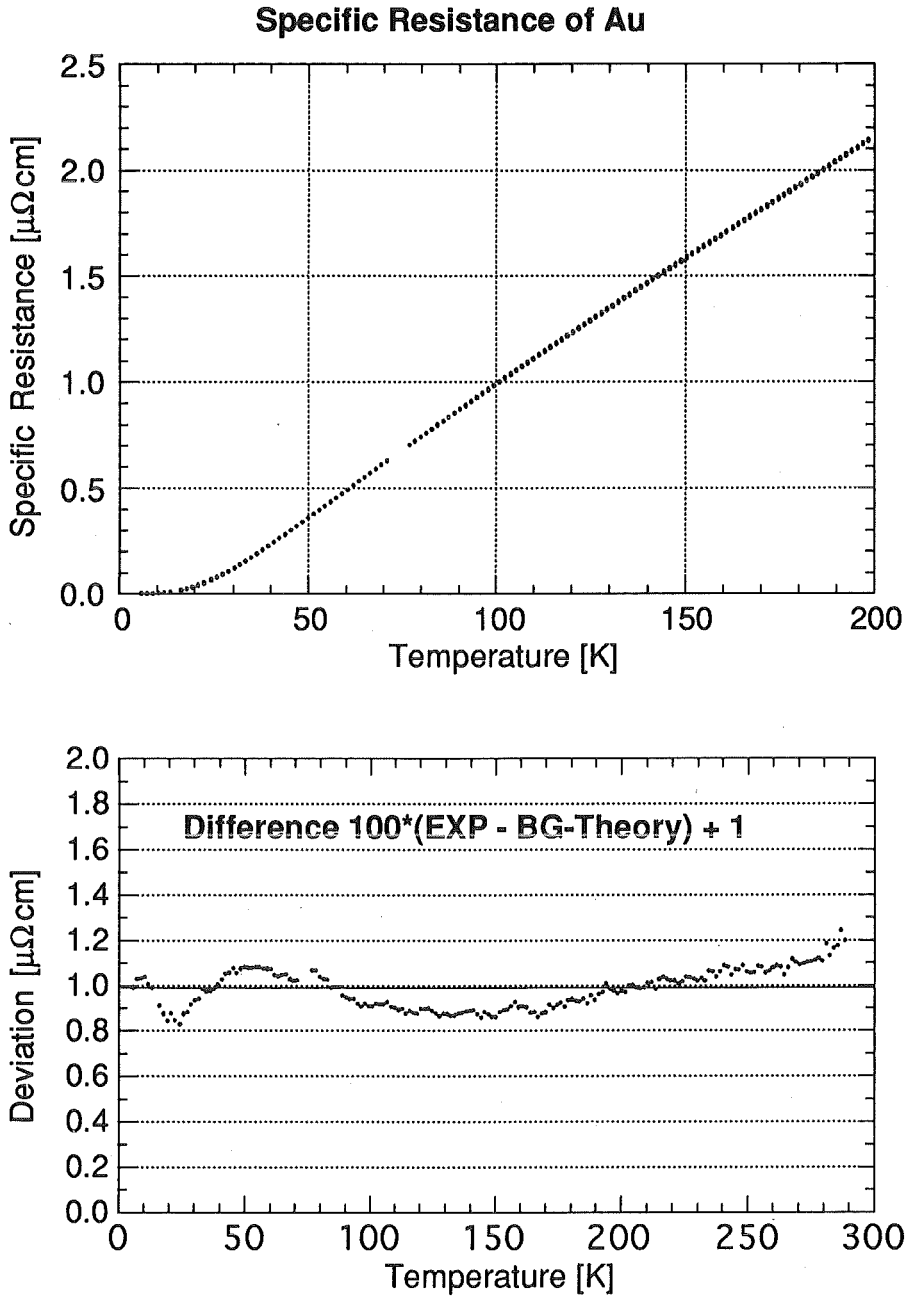

Figure 2. Plot of $\rho(T)$ for a gold film. Data points and the least squares fitted Bloch-Grüneisen theoretical curve lie right on top of each other. On top only the data are shown. A few points were left out, due to electronic malfunctioning. Note the linearity above 170K, and the low temperature $T^{4.0}$ behavior. The negative intercept ρ_o is -5% of $\rho(RT)$ and - 7.5% of $\rho(\Theta_D)$. The bottom figure shows that the experimental data and the fitted theoretical curve deviate at most by 0.2%. Note the factor of 100.

In recent years some absolute and essentially first principle $\rho(T)$ variational calculations have been carried out for metals. See references 21 and 22 as examples. Absolute errors of the order of 10% in the high temperature regions are still obtained, with possible higher discrepancies in the low temperature region. The experimental shapes are well reproduced. These calculations are based on a theory using the electronic density of states and the Eliashberg function $\alpha^2F(\omega)$, reflecting the electron-phonon coupling. It appears, that for the HTSC materials, this theory has to be modified for electron-electron interaction.

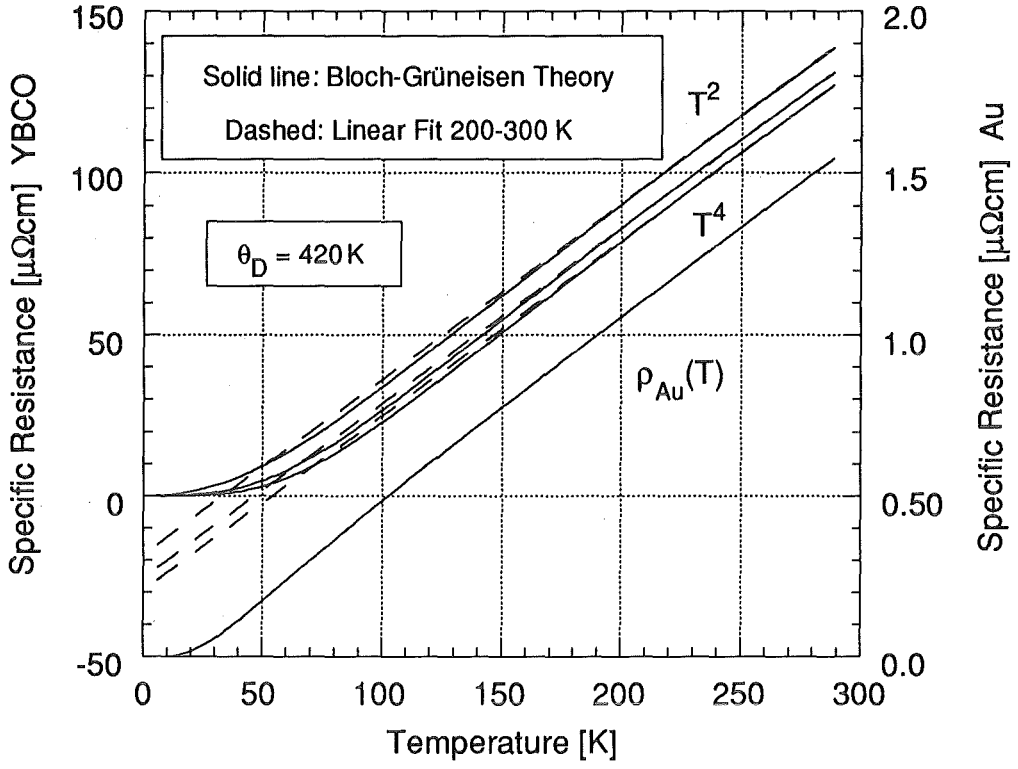

Figure 3. Plot of the Bloch-Grüneisen $\rho(T)$ for $Y_1Ba_2Cu_3O_7$), Θ_D=420K for n=4.1 (T^4 shape), n=3.1 and n=2.1 (T^2 shape), without a superconducting transition. Note that the calculated curves lie below the extrapolated line at intermediate temperatures. Note also that this variation over the naturally occurring range of n has but little influence on the size of the negative intercept ρ_o of -16 to -26% of the RT value, depending on n. Our experience lets us expect a T^4 shape, because of the high phonon cut-off of the cuprates. However decreasing Θ_D to 153K (T^4 shape) decreases ρ_o substantially to -5% of the RT value for Au (right scale). The curves have been scaled to the same slope.

Our best films show a definitely increasing slope above about 170K.

Recently, in pursuing the preparation of well ordered films, Poppe et al. [23] obtained films that no longer show a linear rise above 120K in the $\rho(T)$ curves, but rather an increase in the slope m from T_c to RT, with a maximum of the slope at 230K. As shown in figure 4, the top and bottom curves of their figure 2 can well be fitted with $\rho(T) = \rho_o$+ const. $* T^k$, where k=1.28, and 1.39 for the better bottom curve.

In fact, their sample with the best $\rho(T)$ data, NP09081D, has k = 1.77 [24]. So obviously, as the samples become better in the sense of having fewer defects, the exponent k increases. Sample Sr2803 shows a negative intercept ρ_o of -2μΩcm, when extrapolated from the tangent at 100K and -55μΩcm, when done so from the tangent at 300K. This curve can be fitted with $\rho(T)$=10.7+0.0292$*T^k$ μΩcm, yielding a very low level of residual defects. For a perfect sample ρ_o should be zero of course.

The exponent may well end up at k = 2.0, as observed for some organic superconductors [12], with conduction in two dimensions. Such curves show a quadratic form above 40K, with a shape not quite settled below.

For some organic superconductors [25], having conduction in one and two dimensions, a quadratic dependence on T is observed from 40 to 300K. This also approximately holds in the planes for graphite [26], transition metals dichalcogenites and their intercalated com-

pounds [27]. Hence it is inferred, that Poppe's data indicate the transition of the conductance from three to two dimensions. As the number of defects is sufficiently reduced, the ρ(T) curves should approach a T^2 behavior, as observed for conduction in two dimensions. A similar transition has been observed in $Tl_2Ba_2CuO_{6+\delta}$ [28], although possibly not correctly interpreted. For $Nd_{1.85}Ce_{0.15}CuO_4$ k=2 ± 0.1 has been observed for the in plane resistivity [29]. The existing evidence, together with the observed strong ρ(T) anisotropy and behavior in other layered systems, points overwhelmingly to a T^2 dependence. Such a T^2 shape is difficult to explain for electron-phonon, but naturally holds for electron-electron interaction.

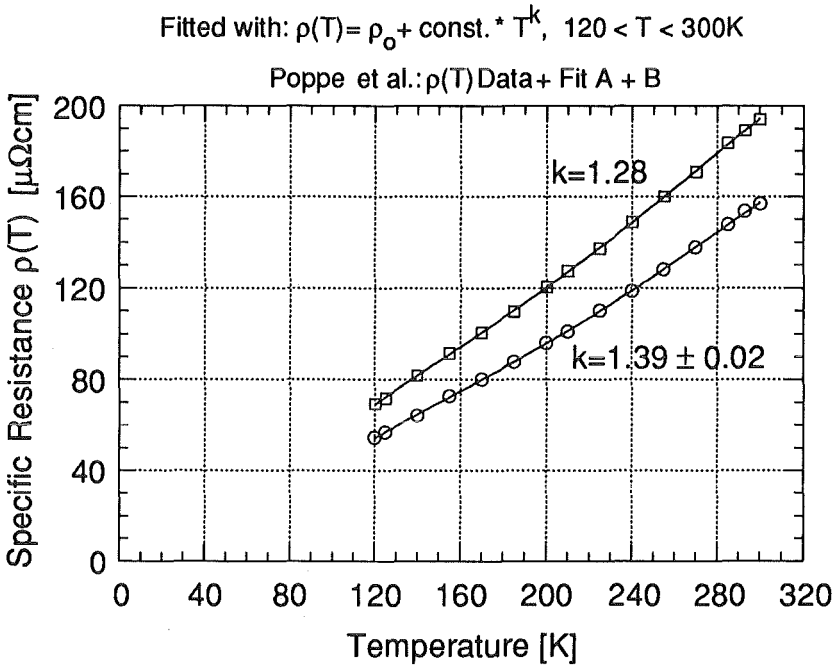

Figure 4. Fits of two of the ρ(T) curves of ref. 23 for $Y_1Ba_2Cu_3O_7$, above the region of fluctuations, without the superconducting transition. The top curve in figure 2 of ref. 23 has k=1.28. The better bottom curve has k=1.39. Their best sample, NP09081D has k=1.77, indicating the transition to conduction in two dimensions, as for some organic superconductors and (intercalated) graphite and transition metal dichalcogenites, like $NbSe_2$.

To the authors knowledge, such a behavior has not previously been observed so clearly in the 123 system, except for a slight indication in the ρ(T) curves of the present data and single crystals [30,31]. The deviation from linearity seems to be most pronounced for the b_o direction, which is the direction of the chains. The a-b anisotropy in ρ(T) is attributed to the chains.

The recent measurements of ρ(T) in single crystals [31] show slopes of $m_a = 0.55$, $m_b = 0.25$ in the Cu planes, and $m_c \geq 12.6 \mu\Omega cm$ /K perpendicular to the pairs of planes. We expect the latter value to rise, on the basis of the extrapolation of past data. For the Bi-systems perpendicular to plane / to in plane resistivity ratios larger than 10^4 have been observed.

This implies that once a very low level of defects has been reached, conduction takes place essentially only in the buckled CuO_2 pair of planes, and there preferentially along the CuO chains. This, however, has the consequence that one of the basic assumptions in both the Bloch-Grüneisen and modern theories [22], namely conduction in three dimensions, is no longer valid. The corresponding matrix element, say formula (18) in ref. 22, has then to be evaluated in two dimensions. For the Bloch-Grüneisen theory [19,20], this modification is a moderate effort, but for the variational methods based on first principals, this requires a

significant programming and computing effort. When done, the integration over the fuzzy (because of the buckling) Fermi cylinder leads to a T^2 dependence for the specific heat, in agreement the data for graphite. For 3D systems T^3 is obtained. This exercise demonstrates the consequences of the reduction from 3D to 2D, but of course only for electron-phonon interaction. The above arguments permit a prediction: The specific heat for 2D superconductors, above T_c , should be proportional to T^2 up to about room temperature, if the Wiedemann-Franz law holds, as stated in ref. 27. That experimentally a T^3 low temperature behavior is observed [32], would then be an unresolved contradiction. However, the calculations in ref. 34 resolve this discrepancy, with a T^3 result.

Just as for graphite [26], the phonon spectra will be highly anisotropic as well. Measurements on polycrystalline samples give averaged values only. Such measurements are then needed on single crystal samples or a least on melt textured ones. Recently such measurements were done on 123 single crystals [47]. The data show anisotropy indeed.

For thin films the highest anisotropy ratio reported [33] to the authors knowledge is 2.6. The discrepancy between single crystal and thin film data implies that no nearly perfect single crystal thin films have been prepared. This basic problem has not been solved yet.

4. Summary and Conclusions

Quality criteria for HTCS, based on the T_c downset and transition width as a function of the substrate temperature, and on slope and negative intercept of the extrapolated linear part of $\rho(T)$ are discussed. Further tools are optical microphotography, Rutherford backscattering and channeling measurements. The 3D Bloch-Grüneisen theory is a further guide.

A high and sharp (<0.2 K) transition temperature is a necessary, but not sufficient criterium on whether a given sample is well-ordered, as this can be caused by a few percent of the material, allowing the first superconducting current path. $\rho(T)$, giving an average value over the whole sample, is a better and more sensitive criterion of atomic ordering. As working hypothesis, it was assumed that well-ordered 123 samples have the $\rho(T)$ shape of a monovalent metal like Cu or Au. Then $\rho(T)$ for 123-samples should be linear above about $\Theta_D/2$ = 210K, up to about 750K, when the oxygen starts to diffuse out. Below it should have the shape given by the 3D Bloch-Grüneisen theory, that is a low-temperature T^n behavior, with $2 < n < 4.1$, corresponding to the range observed so far in nature. As the CuO_2 pair of planes cause a peak around 85mV in the phonon spectrum, the author expects a n value of about 4. Of course it should be born in mind, that the Bloch-Grüneisen theory is based on electron-phonon coupling. For electron-electron coupling in the 3D quasiparticle theory dimensionality arguments yield a T^2 shape above 30K and slight structure below [34]. Additionally there will be an additive temperature independent term ρ_0 caused by defects.

Our 123 films show a variation in the slope m by a factor of 5 and of ρ_0 by a factor of 60, when fitted with $\rho(T) = m*T + \rho_0$. The slope m and ρ_0 as a function of substrate temperature T_S show narrow reproducible minima, whose positions fall with decreasing oxygen pressure. Some of our films show a slope, that increases above about 170K, but with $k<1.1$ for a T^k fit.

The experience with A15 films and the analysis of literature data shows, that only when the preparation parameters are controlled within very narrow "windows", that the atomic ordering processes of nature cause a homogeneous and nearly perfect growth of the samples, with few weak links, where the impurities segregate to. The analysis of our 123 films permit the statements that the substrate temperature has to be controlled to an accuracy of

less than 1K, the stoichiometry to less than 1%, probably < 0.1 at% and the oxygen partial pressure to less than 5%. So it appears that the 123-system is a real line compound. Once sufficient control of the preparation parameters has been achieved, then hopefully the atomic ordering processes (or driving forces) inherent in nature will take over and lead to nearly perfect growth. Reproducibly a $m_a = 0.6$ value was found, in agreement with $m_a = 0.55$ for the best single crystals.

However, a entirely unexpected result is that m and ρ_0 are almost totally correlated, which constitutes a complete violation of Matthiessen's rule. This is presently attributed to percolation [11]. But "We feel that this implies that the mechanism that gives rise to the temperature dependence is the same as the mechanism that gives rise to ρ_0, i.e. elastic scattering by defects. Only when ρ_0 is extremely small, the scattering by phonons" ? "should take over"[35]. A possible explanation is weak links [11].

The 3D Bloch-Grüneisen theory yields as one result, that ρ_0 is negative, with values of -16% (n = 2) and -26% (n = 4) of the RT value for perfectly ordered samples. It is to be expected that the superconductor community will soon have a set of three highly anisotropic, universal $\rho(T)$ values. These values should in good approximation also hold when Y is substituted by other rare earth elements and for systems with more than one pair of CuO_2 planes, depending slightly on Θ_D. This permits quality control for samples with high critical currents, that is many defects and hence a linear $\rho(T)$ shape.

Recently Poppe et al. have produced 123 films with c orientation, whose $\rho(T)$ curves above 120K are no longer linear. They can well be fitted with $\rho(T) = \text{const.} * T^K + \rho_0$, with k up to 1.77 [24]. As single crystal data prove that electrical conduction essentially takes place only in the CuO_2 planes, this was interpreted as the transition of conduction from 3D to 2D. Organic superconductors as well as (intercalated) graphite and transition metal dichalcogenites like $NbSe_2$ have an approximately T^2 behavior in $\rho(T)$. It was generalized that this also holds for HTCS. This holds for $Nd_{1.85}Ce_{0.15}CuO_4$ and other HTCS [45]. An extensive comparison between organic and oxide superconductors has been made by Green [46] and shows numerous similarities in the physical properties. These compounds also show a different low temperature exponent n, in and perpendicular to the planes, indicating anisotropic phonon spectra, as experimentally observed for 123 single crystals [47].

By generalization is meant a working hypothesis that supported by additional experimental evidence or sometimes theory. Once about six pieces of evidence have been obtained, and there are no contradictions, the thesis comes close to certainty. But flaws are not excludable.

Gurzhi et al. [34] have theoretically in a qualitative way calculated physical properties of 2D layered structures in general. The momentum transport of the moving electrons requires, because of momentum conservation, a mechanism to transfer momentum to the lattice (momentum relaxation). As soon as the defect concentration falls below a critical value, the electron-phonon scattering is severely suppressed. Then the momentum relaxation must be accomplished by the weaker electron-electron scattering. This has a T^2 shape in $\rho(T)$ as consequence, in agreement with experiment and as predicted by Ruvalds and Virosztek [36]. While Weger [35] disclaims the validity of Ghurhzi's work for higher Fermi energies, an E_F value of 0.1eV is usually given [46,48], which would be in the range of validity. The Hall effect, related to the off-diagonal elements of the conductivity tensor, is expected to show the same exponential rise with falling T , and does so experimentally over almost three orders of magnitude [12]. For 3D systems $R_H = -1/nec$, independent of temperature, is expected in first order (see Ref. 51, p. 14). This constitutes further experimental proof of an electron-electron superconductivity mechanims, perhaps permitting RT superconductivity.

Further: The missing of the Hebel-Slichter peak is best evidence against a BCS picture [37]. Aronov in a talk at Karlsruhe University (Dec.11, 1991) made the statement that the behavior of the Hall effect of 123 is proof of the non-BCS nature of HTCS [38]. As the phonon-electron scattering is strongly suppressed, a possible explanation is that the mechanisms for HTCS is caused by electron-electron interaction, following the ideas of Landau in the late fifties, but in 2D. By the way, this T^2 dependence can be derived for the 3D quasiparticle theory solely by dimensionality arguments [51]. The HTCS materials are possibly the first systems, where this can be demonstrated. This dimensionality argument appears to hold both for 2D [34] and 3D [52].

Then two electrons condense to a bound boson (bipolaron), with a charge of $2e^-$. Experimental evidence supports this [39]. Numerous arguments supporting s-wave electron-electron pairing have been summarized by Varma [50] and Anderson [54].

The specific heat has, as in 3D, a T^3 shape [33]. The Wiedemann-Franz law predicts T^2 behavior for the specific heat. Ref. 27 however in 2D predicts the observed T^3 shape. Also it is pointed out that both thermal and electric transport conductance are tensors in anisotropic systems.

Further: "The mean free path l is very large; I estimate it at about 250Å at 100K (for m = 0.25), and more at lower temperatures. It is the concentration of defects (such as oxygen vacancies or disorder) so small, that the distance between defects is so large? Or, alternatively, is the scattering between defects suppressed by some mechanism?" [35]. The answer is that the relaxation times grows exponentially as the temperature is lowered - for 2D systems [34].

Further proof is given by the temperature dependence of the thermo-e.m.f. It rises rapidly above T_c and then goes into saturation up to RT [40]. Ref. 34 contains further predictions to be tested experimentally, in particular in sound attenuation. See ref. 43 and other related papers.

It should be pointed out that electron-phonon coupling is suppressed, but not forbidden. This may be the cause for the observed departure [45] from a T^2 shape above 200K. Also in real samples there remain residual defects. Both can provide some electron-phonon contribution.

T_c appears to be enhanced, as the Fermi level is found to lie near an almost logarithmic van Hove singularity in the electronic density of states [48], using the Fermi liquid theory. That paper contains further proof, showing reasonable agreement between experiment and theory.

It should be pointed out, that the bipolaron theory of HTCS superconductivity by Emin [41] predicts a linear $\rho(T)$ behavior in the range $170 < T < 600K$. As the $\rho(T)$ calculations in two dimensions confirm the nonlinear shape, this theory is invalidated and has to be modified or abandoned. Perhaps the only flaw is the calculation in 3D. This also pertains to any other theory predicting such a linear form. So there may be no need to invoke exotic models [12] (p. 2-3). The electron-electron scattering theory [36] does predict a T^2 behavior of $\rho(T)$. However, it should be pointed out that this can probably be accounted for by the conduction in two dimensions in the HTCS systems. See ref. 34. This T^2 behavior does not conform with Weger's linear ρ_{ab} [12]. That model is based on electron-phonon interaction.

Other theories should be checked, to what extend they are effected by the dimensionality arguments. Another possibilities are the marginal Fermi liquid theory [42] and the Luttinger liquid theories, which explain the strong temperature dependence of the Hall ef-

fect [53]. Those theories are 2D and are also supported experimentally. The discrepancy lies in the imposed linear power of $\rho(T)$.

So it appears that theory and experiment finally meet and the gap is closed. Perhaps the reduced dimensionality together with the suppressed electron-phonon scattering permit finally an experimental proof. But maybe it is just the exponential increase in the mean free path as the temperature falls.

The following remarks should be viewed with a grain of salt, as the author has theoretical background, but is not a theorist. Landau's quasiparticle theory is based on electron-electron interaction and has the experimentally required T^2 shape for $\rho(T)$. These theories are usually referred to as Fermi liquid theories. They were modified to account for a linear $\rho(T)$ shape and are then called marginal Fermi or Luttinger liquid theories. They make sensible predictions otherwise (bipolaron model). So it is up to the theorists to see whether it is possible to modify their theories so that they, while keeping the presently good agreement with numerous other experimental results, can obtain <u>one</u> theory that also yields the T^2 shape for $\rho(T)$ .While the T^2 shape of $\rho(T)$ probably provides the crucial experiment to distinguish between electron-electron and electron-phonon interaction, further such (crucial) experiments might then be needed to distinguish between the various versions of the electron-electron models.

5. General Remarks

Ideas and calculations have been presented on how to decide when nearly perfectly ordered samples will have been reproducibly obtained. The strategy proposed is not immediately compatible with optimizing the critical current J_c. Rather, a two step procedure is proposed. The first is to obtain nearly perfectly ordered samples. Then, if necessary, the needed defects should be put in to maximize J_c. That is the strategy that finally succeeded for Nb_3Sn. One should not try to do the second step before the first.

Presently it appears that a sensible strategy is to optimize the exponent of the $\rho(T)$ curves above about 120K and to minimize the critical current J_c, always for single phase films. In addition to the c-axis orientation, the films should also have a-b orientation to reduce the problems associated with weak links. This problem might be solved by using tetragonal substrates with the needed a/b ratio of about 1.01 or perhaps by cutting cubic substrates at a suitable angle of a few degrees. The films then hopefully start growing at the atomic steps of the substrate. Perhaps only the combination of both will work, but such films might have a good structure, but poor transport properties between the layers.

So, in referring to the second opening paragraph, relevant properties to be optimized are the sets of slope, intercept and curvature of the $\rho(T)$ curves. Further one should initially minimize J_c and then, if needed, maximize J_c through controlled addition of pinning centers. Subsidiary conditions are reproducibility and, usually, phase purity.

The whole paper can be condensed into one simple generalization:
High T_c superconductivity is two dimensional in nature, with all ensuing consequences. A generalization is no proof, but it permits testing by predictions and can be checked by existing and doable experiments. The appropriate dimension must be used in theories. At the same time, this dimensionality simplifies many calculations [34].

The author apologizes for not having cited the work of other authors, whose work is relevant. The literature studied and the present list of references was obtained by aimed searches, rather than a comprehensive search of the whole literature.

Experiments that depend on a low concentration of defects, may be invalid, if done on samples not free therefrom, and may have to be repeated.

There is a lesson to be relearned for the HTCS field: If your results are not reproducible, that is natures gentle way of telling you that you should improve you equipment until they are, or that you are not using it properly. Do not ignore that message. Never abandon the postulate of reproducibility. It is one of the fundamental attributes of science. No excuses.

A similar remark applies to some theories. It appears to be possible to twiddle with parameters until all existing data fit. That is no proof of that theory. The author hesitates to take serious any theory that does not make a prediction that can be tested by experiment.

The author is of the opinion that these are basic quality criteria generally applicable in physics.

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