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SUPERCONDUCTING NIOBIUM CAVITIES, A CASE FOR THE FILM TECHNOLOGY

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Abstract

Evidence is presented for niobium film cavities performing as well as niobium bulk cavities, at variance with a widespread belief that their much smaller grain size should be a fundamental limitation preventing high quality factors to be maintained over a wide range of accelerating fields. By comparing the relative merits of the bulk and film technologies, a strong case is presented in favour of the latter.

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1. INTRODUCTION

The successful operation of LEP2 [1] has demonstrated the feasibility of using on a large-scale copper accelerating cavities coated with a thin superconducting niobium film. As many as 272 such four-cell cavities are currently installed in the LEP tunnel and routinely operated at 4.5 K in their fundamental 352 MHz TM_{010} mode. An experience of several years did not reveal any major operational difficulty. On the contrary the average accelerating field has now exceeded the design value by 1 MV/m, thus reaching 7 MV/m, corresponding to an energy of 100 GeV per beam, in rather extreme synchroton radiation conditions (a power of 18 MW for a total energy loss per turn of 3.4 GeV). In spite of this success, and of the well known advantages [2] of the film technology over the bulk technology, other existing or planned installations, such as CEBAF [3] and TESLA [4], rely fully on the latter. The reason is a widespread belief that the film technology should suffer from fundamental limitations preventing large quality factors to be maintained over a wide range of accelerating fields [5,6]. Such a belief has its roots in erroneous interpretations of early data, making no explicit distinction between the residual and BCS components of the surface resistance (the latter is dominant at LEP2) and accepting the conjecture [7-9] that most of the residual resistance of niobium films should be blamed on their small grain size, and therefore could not be reduced to negligibly small values. Now that an extensive set of data [10-14] is available which makes it possible to better analyse and understand the mechanisms contributing significant RF losses in niobium films, such a conjecture is no longer tenable and new light has been shed on the compared merits of the bulk and film technologies. The present paper aims at a brief and simple presentation of the arguments of relevance. After a short review of the main results obtained in recent years, convincing evidence is presented that the film technology allows for as good a performance as the bulk technology. The additional well known merits of the film technology, including the better thermal conductivity, the lower cost, the lesser sensitivity to stray magnetic fields and the simpler manufacturing procedure, are presented next, hopefully making it clear that it should be preferred to the bulk technology in future large scale applications.

The data [10-14] reviewed in the present paper were obtained using single-cell cavities of a geometry similar to that of CEBAF and TESLA cavities, operated in the fundamental 1.5 GHz TM_{010} mode. A detailed description of the experimental method, including the production of 1.5 µm thick films coating the inner wall of copper cavities, the measurement of their surface resistance down to temperatures of 1.7 K, and the analysis of the data in terms of a limited number of variables are available in Reference [10] and are not repeated here.

For the purpose of the present work it should be sufficient to recall that the quality factor Q and the surface resistance R_s are related to each other via the relation $Q = 295\ \Omega / R_s$ while the average RF magnetic field on the cavity surface, H_{rf}, and the electric field gradient along the cavity axis, E_{acc}, are related to each other via the relation $H_{rf}[mT] = 4.55\ E_{acc}\,[MV/m]$.

It is convenient to split the surface resistance in three terms, the BCS resistance R_{BCS}, the fluxon-induced resistance R_{fl} and the residual resistance R_{res}, $R_s = R_{BCS} + R_{fl} + R_{res}$. The BCS resistance accounts for the existence of unpaired electrons when the temperature differs from zero. It is in practice negligible at the superfluid helium temperatures in use at CEBAF and TESLA. To a good approximation its temperature dependence is given by $R_{BCS} \propto exp(-\Delta/T) / T$ with Δ being the energy gap. The fluxon induced resistance [13] measures the sensitivity to external magnetic fields H_{ext} possibly present at the time when the cavity is cooled down below the critical temperature T_c. It is proportional to H_{ext} and increases linearly with the RF amplitude. At 1.7 K $R_{fl}(1.7K) = (R_{fl}^0 + R_{fl}^1\, H_{rf})\, H_{ext}$, the temperature dependence being measured by the variable $k_{fl} = R_{fl}(4.2K) / R_{fl}(1.7K)$ and being much smaller than in the case of the BCS resistance. Finally the residual resistance [11,12] is the only term which subsists at zero

temperature when $H_{ext} = 0$. In practice it is the only harmful term for cavities operated under conditions similar to those in use at CEBAF and TESLA. It is often observed to increase with H_{rf}, first linearly and then, beyond some field emission threshold H_{fe} [15,16], exponentially. For $H_{rf} < H_{fe}$ it is usually possible to write $R_{res} = R_{res}^0 + R_{res}^1 H_{rf}$. As shown in Section 3 both R_{res}^0 and R_{res}^1 can be kept under sufficient control and maintained at low values, similar to those obtained with bulk niobium.

2. MAIN RECENT RESULTS: A SUMMARY

Standard films, such as those coating LEP2 cavities, have been extensively studied. They are 1.5 μm thick niobium films grown by magnetron sputtering (≅ 360 V) in argon atmosphere (≅ 1.5×10^{-3} mbar) on oxidised copper substrates kept at ≅ 150ºC. The gross features of the data [10] are well described by BCS theory, the London penetration depth λ_L, the BCS coherence length ξ_0 and the strong coupling parameter $\alpha = \Delta/T_c$ taking values in good agreement with the bulk values available in the literature.

The critical temperature is about 0.25 K higher for standard films than for the bulk, a result of the higher stresses present in such films. Most of the differences between the superconducting properties of standard films and those of the bulk are accounted for by different electron mean free paths, that of the film taking a value $l_0 \simeq \xi_0 \simeq 30$ nm in good agreement with the measured residual resistivity ratio, *RRR* ≅ 10 to 20. Non-standard films have been produced by changing some of the sputtering parameters (*e.g.* the nature of the substrate, that of the noble gas, etc…). As they span a wide range of mean free path values (and accordingly of *RRR* values), they allow for a more stringent confrontation with the predictions of BCS theory and a good agreement is generally found. In particular the BCS prediction of a minimum of the BCS resistance below $l = \xi_0$ is verified (Figure 1a), illustrating, and at the same time explaining, the large ratio between the bulk and film values of the BCS resistance (in excess of a factor 2). An important and sometimes overlooked observation is the slow rise of the BCS resistance with H_{rf}, typically by 50% when H_{rf} reaches 32±5 mT. This result applies to both the bulk and film cases (Figure 1b), implying a larger effect in absolute terms for the bulk than for the film. To the extent that the characteristic scale is in the range of the critical fields, this result is not surprising.

Less expected is the evidence for spectacular differences between films grown on oxidised and oxide-free copper substrates [14]. In general, the latter have superconducting properties approximately half way between those of the bulk and those of standard films. In particular their critical temperature is about two tenths of a Kelvin lower than that of standard films and their electron mean free path is about twice as large (Figure 2a). These results are related to the lower stresses existing within films grown on oxide-free copper, as has been measured from X-ray diffraction spectra which also provide evidence for an important difference of texture (Figure 2b). Another notable difference concerns the size of the grains, an order of magnitude larger for films grown on oxide-free copper than for films grown on oxidised copper.

The sensitivity of the residual resistance to trapped magnetic flux [13], measured by R_{fl}, is nearly two orders of magnitude larger for the bulk than it is for standard films. This has been described [6] as being the result of an anomalous behaviour of the upper critical field H_{c2}. Recent H_{c2} measurements [13] do not support this conjecture, the results being instead in conformity with what may be expected from a reasonable *l*-dependence of the upper critical field and leaving no room for any kind of exotic behaviour. The properties of R_{fl} are best understood in terms of pinning, apparently dominated by "bubbles" of the noble gas atoms used in the sputtering discharge. Figure 3 illustrates the main features of the data. The strongest effect is obtained for krypton on oxidised copper, with R_{fl}^0 and R_{fl}^1 as low as 3 nΩ/G and 0.4 nΩ/G/mT respectively.

Finally the residual resistances [12] have been shown to be dominated by macroscopic defects such as those resulting from the roughness or from the defective polishing of the substrate. Other potential contributors such as the oxidation of the film surface, the formation of hydride precipitates and the contamination by noble gas atoms have been studied in detail and found relatively harmless to the extent that one knows how to keep them under good control. The current state of the art is reviewed in the next section.

3. FILM RESIDUAL RESISTANCES: THE STATE OF THE ART

Indications [10,17] that very low residual resistances can occasionally be maintained over a wide range of accelerating gradients have been available for some time. Such data have provided early but strong evidence against the extension to niobium films of weak link models [7-9] such as those which describe granular high T_c cuprates [18]. Here we concentrate on a set of measurements [12] obtained with krypton as discharge gas and using electropolished copper substrates. The production conditions are otherwise the same as described in Reference [10]. However, particular attention has been paid to prevent events which might create macroscopic film defects. In particular the sputtering and rinsing systems have been slightly modified to better ensure that all steps following the preparation of the copper substrate are performed in dust free conditions. All films grown on electropolished substrates have low values of both $R_{res}{}^{0}$ and $R_{res}{}^{1}$ (Figure 4). They include films grown on oxidised copper as well as films grown on an oxide-free copper underlayer. At variance with earlier data [10], collected at a time when the values taken by the residual resistance were not yet under sufficient control, the present data demonstrate that low R_{res} values, similar to those obtained in the bulk case, can now be achieved with extreme reliability.

Only a fraction of the data displayed in Figure 4 were obtained with the upgraded rinsing installation. These reached higher accelerating gradients, in excess of 20 MV/m, with quality factors exceeding 10^{10} at 10 MV/m and 5×10^{9} at 20 MV/m (Figure 5). The data collected earlier were usually limited by field emission to accelerating gradients ranging between 10 and 15 MV/m. The extension of this limit to higher values revealed a new limitation around 18 MV/m caused by multipacting. Minor changes in the cavity geometry made it possible to overcome this new limit and to reach accelerating gradients in excess of 22 MV/m. There is no reason to suspect the existence of any fundamental limitation which could prevent reaching even higher gradients if yet cleaner conditions were achieved in the production process.

4. FILM AND BULK TECHNOLOGIES: RELATIVE MERITS

Figures 4 and 5 are spectacular demonstrations that niobium purity is not an important contribution to the residual resistance of a cavity, and therefore is not an essential parameter among those which govern the quality of its performance. No significant difference is detected between the residual resistance of films grown on oxidised copper (*RRR* ≅ 15), films grown on oxide-free copper (*RRR* ≅ 30) and bulk niobium (*RRR* > 100). One might even argue that the first of the three, that having the lower purity, should be preferred in practical applications because of its lesser sensitivity to stray magnetic fields and of its lower BCS resistance. Yet, progress in the bulk technology has been largely governed by the ability to produce niobium of very high purity. The main reason is the need for an excellent thermal conductivity in order to avoid quenches, a requirement which is very demanding in the bulk niobium case and which is automatically fulfilled by the copper substrate in the film case. Another, more anecdotic illustration that niobium purity is somewhat irrelevant is the occurrence of hydride precipitation, the so-called “hydrogen disease” [19], in high purity niobium and its suppression by defects acting as trapping sites for the dissolved hydrogen atoms. The decoupling of the requirements on thermal

conductivity and on superconducting properties is, of course, the main and best known asset of the film technology. The need to reach very high niobium purity is a very heavy constraint on the bulk niobium technology, implying a severe selection of the raw material, in particular a difficult and thorough search for possible clustered impurities such as tantalum, and even implying in some cases annealing at high temperatures. On the contrary the procurement of copper having adequate thermal conductivity is not a problem in the film case. However, in both technologies the need to obtain high quality surfaces is essential and is a major complication in the manufacturing process. This constraint is somewhat reduced in the film case where it is the quality of the substrate surface, rather than the quality of the final niobium surface, which needs to be optimised.

Another strong argument in favour of the film technology is the much smaller quantity of niobium required for the construction, less than one part in 2000. This factor applies directly to the niobium cost, with the practical result that it is a negligible component of the total cost in the film case and a major one in the bulk case. This asset of the film technology becomes particularly important in the case of very large-scale projects, where the mere procurement of very large quantities of niobium is a problem in itself.

The advantage of the film technology over the bulk technology in terms of sensitivity to stray magnetic fields is somewhat fortuitous. It is indeed fortunate that the unavoidable contamination of a film by atoms of the noble gas used in the sputtering process contributes virtually no residual resistance and is at the same time an extremely efficient pinning agent. However, in large gradient applications, it is R_{fl}^{1} rather than R_{fl}^{0} which is determinant in the film case. In the case of strongest pinning (krypton and oxidised copper substrate) it contributes ≅ 45 nΩ/G at H_{rf} = 25 MV/m. This implies that the stray magnetic field should not exceed 0.5 G or so for unshielded operation to be acceptable, a stringent but feasible requirement. In the bulk case the R_{fl}^{0} contribution alone is prohibitive and unshielded operation is excluded.

Finally two other arguments are often mentioned in favour of the film technology, possibly of lesser practical importance: one is the ability to strip a defective film and to reuse the same substrate for growing a new film, the other is the possibility to use other materials than niobium, such as NbTi, NbTiN, Nb_3Sn or even high T_c cuprates. The current state of the art in growing films other than niobium still suffers severe limitations in reaching low enough residual resistances but it is not unreasonable to believe that an adequate R&D effort could overcome the present difficulties.

The above list of assets of the film technology leaves little doubt about its superiority once the earlier suspicions concerning its ability to produce low residual resistances over a wide range of microwave amplitudes have been shown to be unjustified. It should therefore be clear by now that the film technology should be preferred to the bulk technology for future large scale projects. However, the present study was only meant to be a proof of feasibility in favour of the film technology, and in this sense it has been a success, but by no means could it be the final word. Additional R&D effort would be mandatory in order to develop and optimise the manufacturing procedure on an industrial scale in order to guarantee the very high cleanliness conditions which are required and, maybe more importantly, in order to achieve significant progress on the procedure used to prepare the copper cavities to be coated. Now that the importance of the surface quality of the copper substrate has been clearly demonstrated one would be tempted to think that spinning, which is causing such important damage to the copper sheet, may not be the ultimate solution. In particular electroforming, which has been only briefly explored in the present study, should be the subject of detailed studies. More generally, the use of other possible substrates, such as aluminium, and of other possible manufacturing procedures should be seriously investigated.

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Figure 1a - The 4.2 K BCS resistance of niobium films as a function of the mean free path dependent quantity $1+\pi\ \xi_0 / 2\ell$. The high point on the clean limit side is for bulk niobium. The line is the BCS prediction.

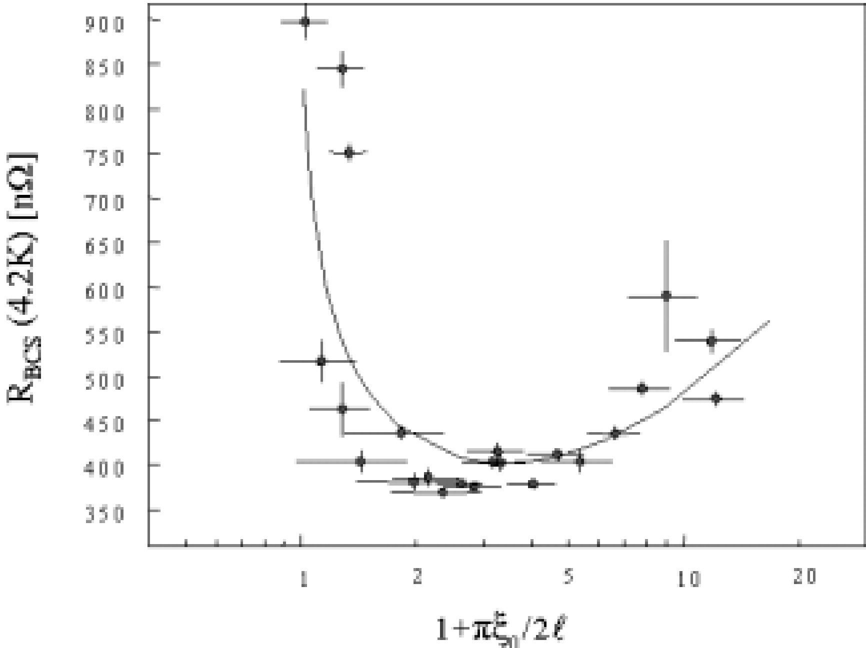

Figure 1b - The 4.2 K BCS resistance of standard niobium films (lower set of points) and of bulk niobium (upper set of points) as a function of the microwave amplitude.

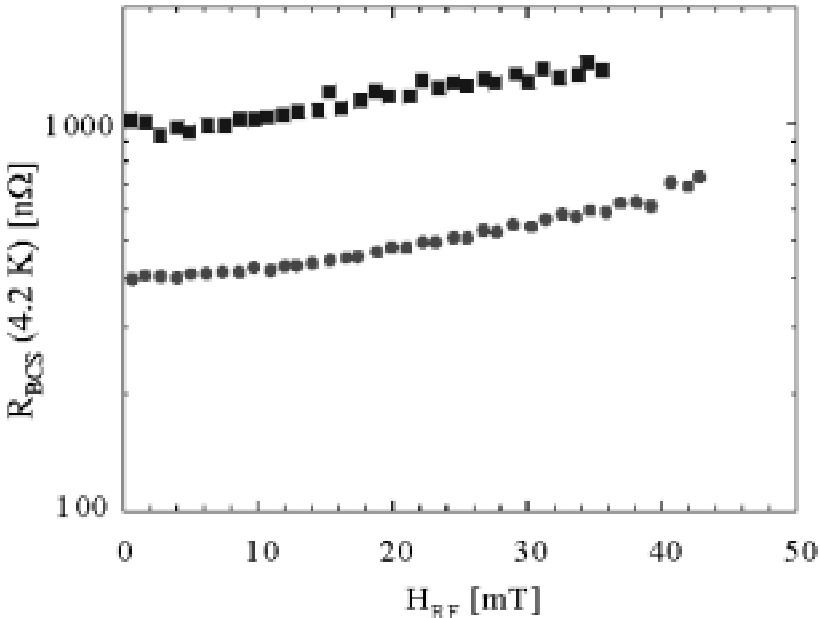

Figure 2a - Distribution in the λ *vs.* T_c plane of different films grown on different substrates in argon, krypton or xenon. Open (full) circles are for films grown on oxidised (oxide-free) copper. The crosses indicate average values.

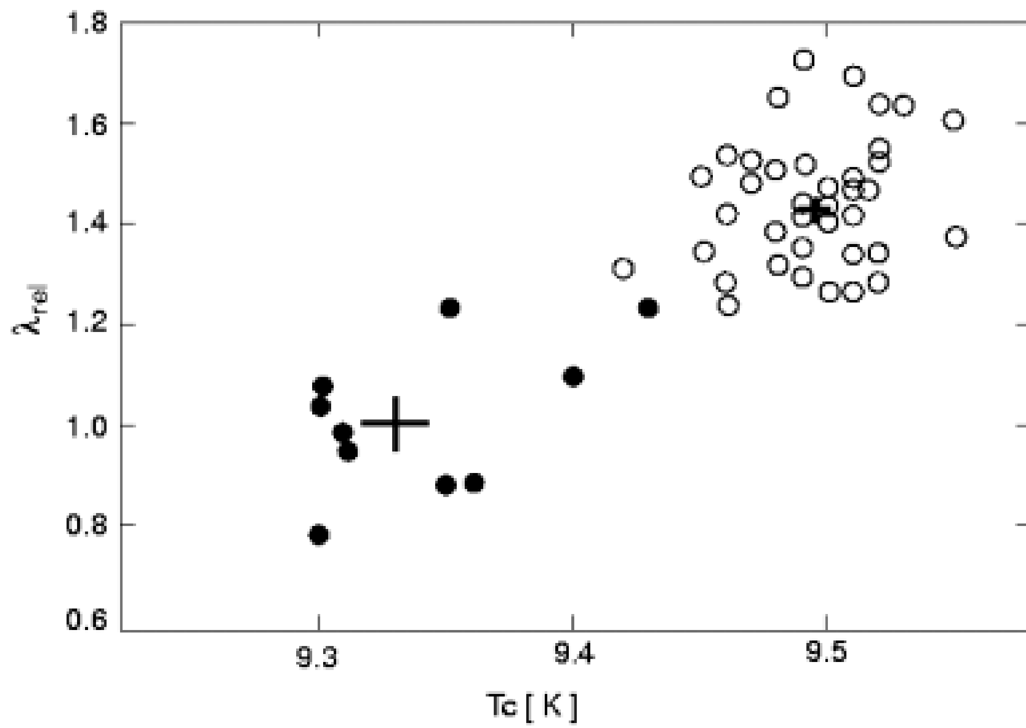

Figure 2b - Bragg-Brentano spectra of equatorial samples of cavities coated on oxide-free (A) and oxidised (B) copper.

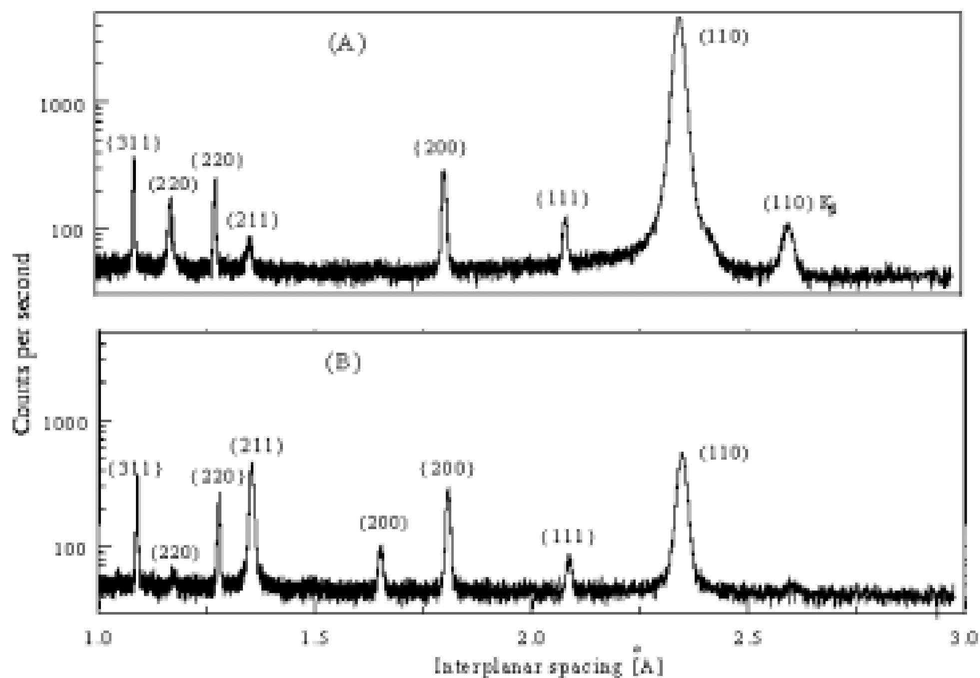

Figure 3a - Distribution of different films in the R_{fl}^{1} *vs* R_{fl}^{0} plane. Open symbols are for films grown on oxide-free copper and full symbols are for films grown on oxidised copper. Data points labelled with numbers are films grown using an argon-neon mixture for sputtering, the label indicating the percentage of argon in the mixture. The line passing through the data points and going from dirty films (top right corner) to the clean bulk limit (cross) via the krypton minimum (left bottom corner) is hand-drawn to guide the eye.

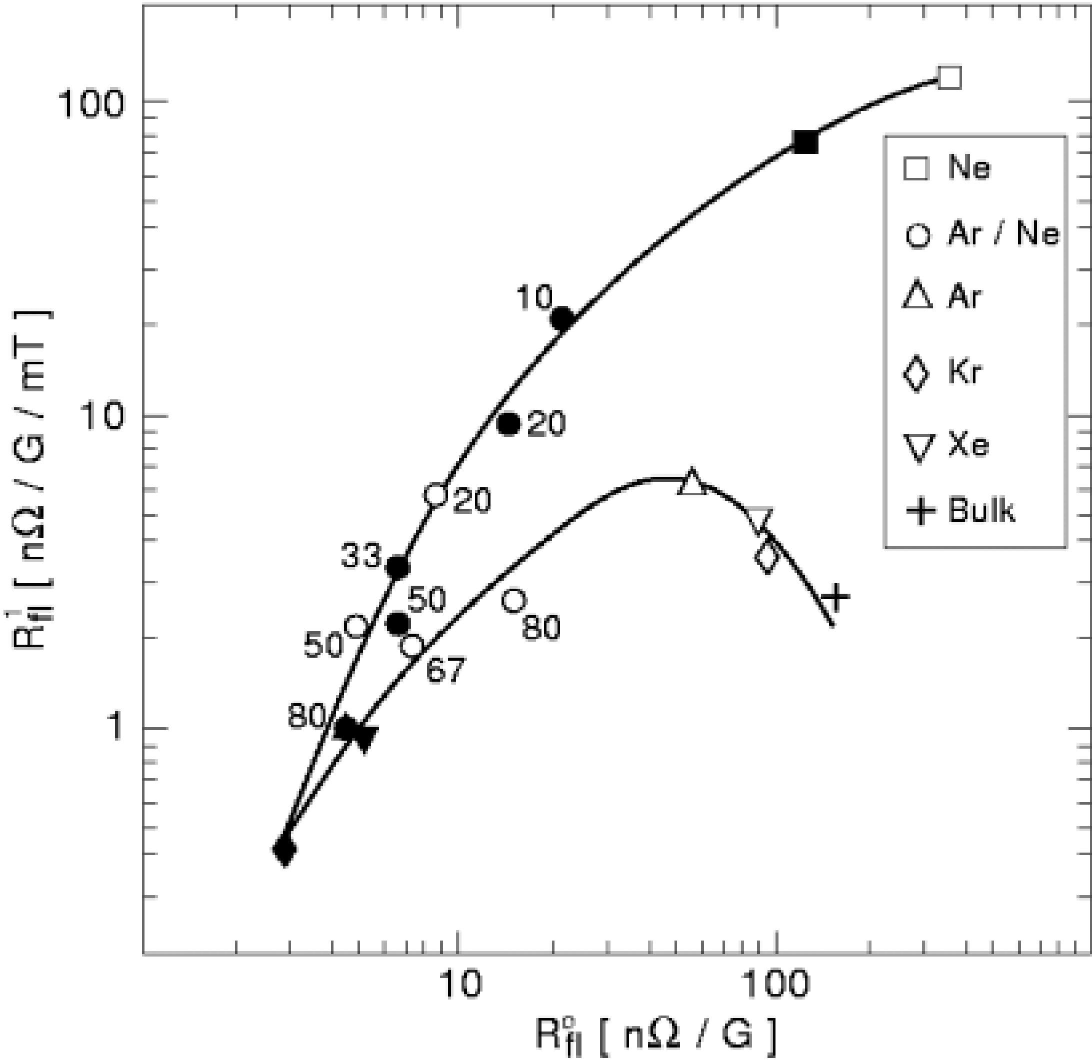

Figure 3b - Distribution of different films in the R_{fl}^{1} *vs* R_{fl}^{0} plane. Argon is used as sputtering gas in all cases and the films differ by the nature of the substrates onto which they have been grown. Films loaded with hydrogen are included as well.

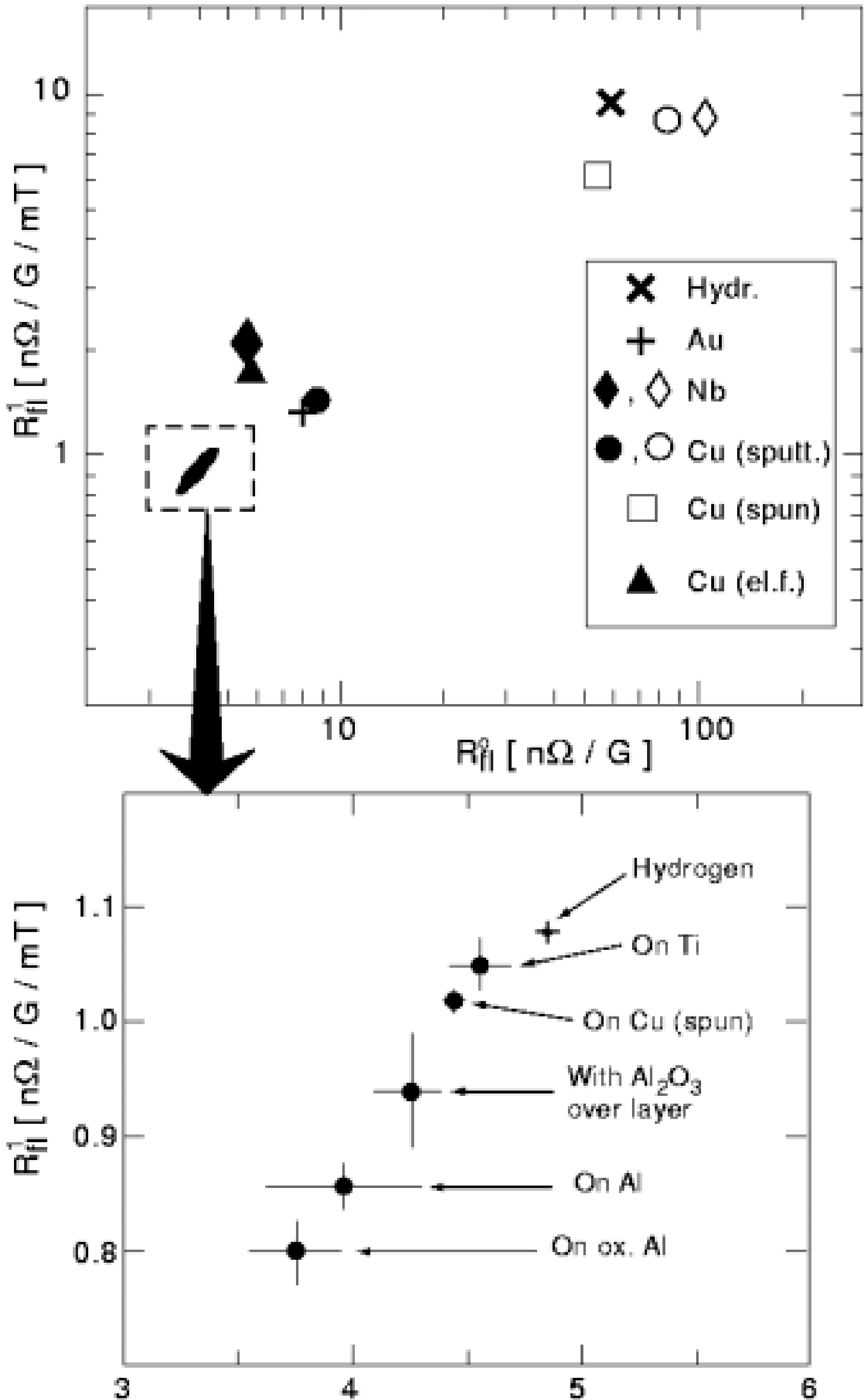

Figure 4 - Distribution of different films in the R_{res}^{1} *vs* R_{res}^{0} plane. The label indicates which noble gas was used for sputtering. Shown are films grown on hydroformed chemically polished copper (squares), on spun chemically polished copper (circles) and on spun electropolished copper (diamonds).

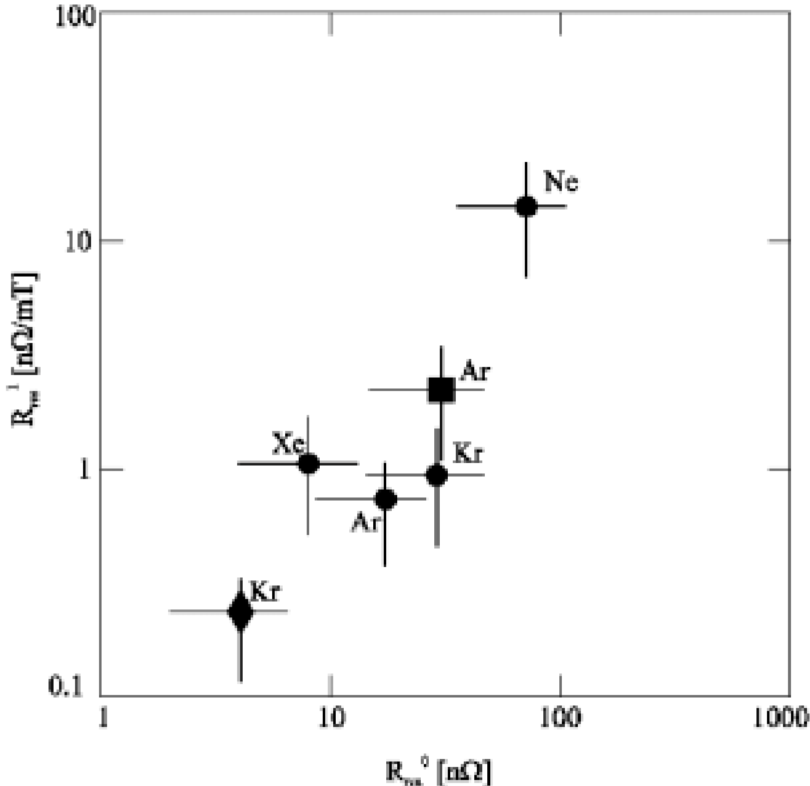

Figure 5 - The quality factor Q of films rinsed with the upgraded installation as a function of the accelerating gradient E_{acc}. All films were produced with krypton as sputter gas.