FLUXON PINNING IN NIOBIUM FILMS

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Abstract

Resistive losses induced by the presence of trapped magnetic flux in niobium superconducting films have been studied using 1.5 GHz microwaves. They are measured to span a very broad spectrum depending on the film-substrate interface and on the gas used in the sputtering discharge. An interpretation in terms of pinning by noble gas clusters is considered.

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1. INTRODUCTION

When a superconducting cavity is cooled down below the critical temperature T_c in the presence of an external magnetic field H_{ext} , the magnetic flux is usually observed to be fully trapped in the cavity walls. In particular this was observed [1] to always occur when using sputtered niobium films or bulk niobium of not too high a residual resistivity ratio (RRR). This feature allows for a measurement of the radiofrequency losses induced by the presence of trapped flux in the cavity walls. The study reported in Reference [1] used 1.5 GHz microwaves at reduced temperatures $t = T/T_c$ typically ranging between 0.2 and 0.5 with values of H_{ext} not exceeding 1 mT. At $T = 1.7$ K the additional surface resistance induced by the presence of trapped magnetic flux can be described by a form:

$$R_{fl}(1.7\text{ K}) = (R_{fl}^0 + R_{fl}^l H_{rf}) H_{ext} \quad (1)$$

where H_{rf} is the amplitude of the magnetic component of the microwave (parallel to the cavity surface). A small temperature dependence was observed which was characterised by a single parameter, $k_{fl} = R_{fl}(4.2\text{ K})/R_{fl}(1.7\text{ K})$, typically ranging between 1 and 3.

Deviations from Equation (1) were observed in several instances, in particular for films having values of R_{fl}^0 not exceeding a few nΩ/G. In such cases R_{fl}^l would often jump to a higher value when H_{rf} would reach a value H_{kink} , typically a few tens of mT, and R_{fl} would not deviate from zero until H_{ext} would exceed a threshold H_{thr} , typically a few hundredths of a mT, above which H_{ext} needs to be replaced by $(H_{ext} - H_{thr})$ in Equation (1).

It has been known for a long time [2] that R_{fl} in niobium takes much lower values for typical films than for the bulk. As most models of fluxon dynamics [3, 4] predict $R_{fl} \cong R_n H_{ext}/H_{c2}$, R_n being the value of the surface resistance in the normal conducting state and H_{c2} the upper critical field, this difference between films and bulk has been blamed [5, 6] on a similar difference between the values taken by H_{c2} . In Reference [1] R_{fl} was measured for a large variety of films spanning a wide range of electron mean free paths and the evolution of R_{fl}^0 and R_{fl}^l was tracked for the first time from the dirty limit (films containing more than 10 atomic percent of neon) to the clean limit (deep-drawn bulk niobium subsequently annealed at 1100°C). Both R_{fl}^0 and R_{fl}^l were observed to display a steep minimum when the electron mean free path ℓ approximately matches the BCS coherence length, $\xi_0 \cong 33$ nm. This result casts some doubt on the validity of the commonly accepted explanation [5, 6], in particular as an independent measurement of H_{c2} [7] was in conflict with earlier measurements used in the argumentation developed in Reference [6]. It was instead proposed in Reference [1] that pinning should provide the dominant contribution to R_{fl} . In particular the possibility that the noble gas used in the sputtering discharge could condense into bubbles, as it is known to do under slightly different experimental conditions [8-16], was considered as a possible source of efficient pinning [17].

In the present work, we report new measurements providing additional evidence for the picture sketched in Reference [1] and we analyse in more detail the properties of R_{fl} .

2. THE UPPER CRITICAL FIELD

The value taken by the upper critical field is essential to the understanding of the fluxon structure and of their dynamics [3, 4]. Accurate measurements are available for pure niobium [18–20] showing that for $t \leq 0.5$ the temperature dependence of the upper critical field H_{c2} and of the thermodynamical critical field H_c agrees to within a few percent with the Gorter-Casimir expressions

$$H_{c2}(t) = H_{c2}(0) (1 - t^2) / (1 + t^2)$$

$$H_c(t) = H_c(0) (1 - t^2).$$

To a good approximation, $H_{c2}(0) = 0.40$ T and $H_c(0) = 0.20$ T. In the presence of impurities H_{c2} is expected to increase as ℓ decreases [3, 4]. Extensive data [21] are available on the increase of H_{c2} resulting from the presence in the niobium lattice of interstitial oxygen in various proportions. They show that H_{c2} is multiplied by 1.54 (resp. 2.17) when the RRR value is reduced to 17.6 (resp. 6.3). Quantitatively, the H_{c2} vs ℓ relationship may of course depend on the detailed mechanism responsible for the reduction of ℓ and the above result cannot be naively extended to the films under study in the present work. It only gives an idea of the kind of increase to be expected and cannot exempt us from performing direct H_{c2} measurements.

We reported earlier [1] on H_{c2} measurements performed on so-called standard films, *i.e.* niobium films, 1.5 μm thick, sputter-deposited on an oxidised copper substrate at $\approx 150^\circ\text{C}$ in argon atmosphere. Two samples of such films were measured inductively at Wuppertal University after dissolution of the copper substrate in a nitric acid solution [7]. The result, $0.45 \text{ T} < H_{c2} (4.2 \text{ K}) < 0.85 \text{ T}$, was much lower than obtained from earlier measurements performed in the presence of the copper substrate [22]. It gave no support to the conjecture that very large values of H_{c2} (in excess of 10 T) could explain the low values taken by R_{fl}^0 in standard films [6] and clearly demonstrated the difficulty of performing reliable inductive measurements of H_{c2} in the case of niobium films kept on their substrate.

It was therefore decided to evaluate H_{c2} from magnetisation measurements, which are known to be of an easier interpretation. Samples were cut from two cavities coated with 1.5 μm films sputter-deposited at 150°C in argon, one (A) on oxide-free copper and the other (B) on oxidised copper. Some of the parameters characterising the two films are listed in Table 1 and reveal important differences. In particular R_{fl}^0 is 17 times larger for A than for B. These differences are known [1] to be related to different textures and stresses, they are discussed in more detail in Reference [23].

The H_{c2} measurements were performed at Vienna University using a SQUID magnetometer equipped with a 1 T magnet. Two sets of measurements were taken, one by lowering the temperature from above $T_c(H)$ for a fixed field H , the other by varying the magnetic field for a fixed temperature T . In both cases the value of H_{c2} could be defined unambiguously and good agreement was found between both sets of measurements (Figure 1). A temperature dependence of the form $H_{c2}(t) = H_{c2}(0) (1 - t^{1.7})^{1.1}$ is observed to give a good fit to the low t data with the result that $H_{c2}(0) = 0.77 \pm 0.01$ T for A and 1.150 ± 0.025 T for B. These values are consistent with expectation and are in a ratio ≈ 1.5 instead of 17, clearly excluding the possibility of blaming the difference observed in R_{fl}^0 on a similar difference of H_{c2} .

In the case of sample B, measurements were made both with the film kept on the copper substrate and with the film removed from it (Figure 1). No significant difference was observed

between the two sets of measurements, giving evidence for H_{c2} being independent from stresses but dependent on mean free path, *i.e.* essentially on the different argon contents of the two films, which, as shown in Reference [23], are the result, and not the cause, of the different stresses generated when the films were grown.

3. NOBLE GAS BUBBLES AS PINNING CENTRES

The present work includes the study of a large number of copper cavities sputter-coated with niobium films prepared under various conditions, part of which has been reported earlier [1]. A notable result, illustrated in Figure 2, is the broad spectrum spanned by R_{fl}^0 and R_{fl}^I and the fact that they take values depending essentially on only two factors: the nature of the noble gas used in the sputtering discharge and the nature of the interface between the film and the substrate. As these factors are varied, the data follow a path in the R_{fl}^0 vs R_{fl}^I plane starting at very large values of both parameters for dirty films grown with neon as discharge gas and ending at large R_{fl}^0 but moderate R_{fl}^I values for clean bulk cavities. In-between, R_{fl}^0 and R_{fl}^I have a sharp minimum for films grown with krypton as discharge gas. When the substrate is oxide-free copper the data are shifted towards the bulk end point from the values they take in the case of an oxidised copper substrate. Changing the nature of the discharge gas results in a change of its concentration inside the film, lighter gases corresponding to higher concentrations, and therefore to lower values of the mean free path ℓ [1]. Changing the nature of the film-substrate interface from oxidised to oxide-free copper results in a change of the film texture associated with lower stresses [23] and results in an overall shift towards their bulk values of all the parameters which characterise the film properties (see Table 1).

Before elaborating on the data displayed in Figure 2, let us briefly review the evidence against any significant relevance of other factors. Figure 3 displays the R_{fl}^0 and R_{fl}^I values taken by different kinds of films grown with argon as discharge gas, including:

- films grown on various substrates (niobium, copper, aluminium, titanium and gold, the first three being also used after having been oxidised),
- a film coated with a thin layer of alumina replacing the usual niobium oxide layer[24-27] in order to check on a possible pinning action of the latter,
- films loaded with hydrogen in order to check on a possible pinning action of hydride precipitates [28, 29].

Figure 3 shows that the data cluster in one of two families, depending solely on the nature of the substrate: oxidised copper, oxidised niobium, titanium, gold and aluminium (oxidised or not) populate one family, oxide-free copper and niobium populate the other. Loading the film with hydrogen or replacing the superficial niobium oxide layer by alumina keeps the films in their original family. When looking closely within each family, small but significant differences can be seen. In particular the detailed nature of the copper substrate is a parameter of relevance. Spun copper sheets, as used for films A and B, give lower R_{fl} values than other copper substrates (electro-formed copper in family B, sputtered copper underlayers or oxide-free copper prepared by sputter-etching [1] in family A). However, the spread of the data within each family is much smaller than the separation between the two families and such small differences may be ignored when discussing the gross features of the data.

If pinning is the relevant mechanism causing the suppression of R_{fl} in standard films, these results would suggest that pinning by hydride precipitates, by superficial niobium oxide or by impurities at the film-substrate interface are relatively unimportant while pinning by the noble gas contained in the film would be the dominant process. Keeping in mind the differences of textures and stresses measured between films A and B, it is tempting to globally extend this result to the two families, one (B) having a typical fibre texture, the other (A) being modified by

hetero-epitaxy [23]. Such stresses would then govern the details of the gas trapping configuration inside the film, which, in turn, defines the pinning properties.

The quantity of noble gas contained in the films has been measured from its release when the films are melted. In order to lower the melting point the films were placed in contact with a nickel foil and heated to form a eutectic. When the temperature reaches the melting point (1170°C) the quantity of gas released is measured. Film B was found in this way to contain nearly twice as much argon as film A (Table 1).

The noble gas contained in the films can be expected to condense, at least partly, into “bubbles” (in fact monocrystals) having typical dimensions of a few nm, as suggested by data obtained with implanted ions [8-12]. Such “bubbles” are expected to be efficient pinning centres and have been studied as such in various amorphous films such as Nb₃Ge and Mo₃Si [13-16]. In the present data the lowest R_{fl} values are obtained by sputtering in krypton on oxidised copper. It should be noted that krypton and niobium atoms are of a similar size and that for a bubble diameter of 3 nm the average distance between krypton bubbles, measured over one penetration depth, would then be of the order of 250 nm, which is the distance between neighbour fluxons for $H_{ext} \cong 1.5$ G.

Direct observation of noble gas precipitates would clearly give more confidence in the validity of the pinning picture being explored here. An attempt at such a direct observation was made by transmission electron microscopy studies performed on samples of films A and B, both in plane view and in cross section. However, defects produced by the preparation of the specimen prevent the observation of features on such a small scale.

4. DISCUSSION OF THE RESULTS

Most models of fluxon dynamics [3, 4] follow the pioneering work of Bardeen and Stephen [30] which describes the loss mechanism as resulting from a viscous movement of the fluxons. However, the amplitude of the movement of strongly pinned fluxons, as the present study has been dealing with, is expected to be strongly reduced with respect to that of the more commonly studied configurations. Here the fluxon array is relatively diluted, each fluxon may be expected to be individually pinned and the magnetic field present at cool-down is well below the lower critical field. Most studies available in the literature deal instead with dense fluxon arrays in the mixed phase, often near the upper critical field, and pinned collectively rather than individually. Care must be taken when discussing the results of the present work to keep in mind these important differences.

The particularly simple and ideal case of a single fluxon trapped in a cylindrical hole of radius r [31] may in fact be closer to the present configuration than that depicted in many other much more sophisticated models. The fraction of magnetic flux trapped in the hole induces no additional loss and its complement, the fraction of magnetic flux penetrating the superconductor from the hole boundary, contributes exclusively to R_{fl} . As the scale of the fluxon core is measured by the coherence length ξ , one would expect large values of r/ξ to be associated with small values of R_{fl} and large values of H_{thr} . In real life, the situation is much less simple and a realistic description needs to take into account the nature of the pinning centres (*e.g.* insulator or normal conductor), the relative importance of individual and collective pinning (*i.e.* the relative densities of the fluxon lattice and of the pinning centres) and the distribution of the pinning centres in the direction normal to the film surface, keeping in mind the small size of the penetration depth with respect to film thickness. Moreover, while non-superconducting defects responsible for the generation of a non-zero residual resistance [32] are not expected to contribute significantly to R_{fl} (otherwise a correlation would be observed between R_{fl} and R_{res} , which is not the case) they may play an important role in the understanding of H_{thr} .

Several authors (representative studies are available in References [33-35]) have discussed pinning configurations of some relevance to the present work, but very little quantitative information is available in the literature. To our knowledge, there exists no quantitative evaluation of R_{fl} which could apply directly to the present work and there exists no discussion, and *a fortiori* no calculation, of the dependence of R_{fl} on the amplitude of the microwave field. One reason is the very small amount of experimental information available, the work reported in Reference [1] was first to identify and define R_{fl}^1 explicitly and to offer a comprehensive study of the properties of R_{fl} in the particular and unconventional pinning configuration characteristic of the present work.

A microscopic description of the radiofrequency losses induced by the presence of strongly pinned fluxons presupposes the availability of a microscopic description of the losses induced by the pinning centres in the absence of pinned fluxons. We remarked in Reference [32] that this was not readily available in the literature and would be likely to imply an extension to microwaves of the description given by McMillan [36] of the proximity effect. We argued [32] that, qualitatively, one might expect the proximity effect, which smears the superconductor-normal conductor boundary around a defect over a distance having the coherence length as a scale, to cause the area of the region containing unpaired electrons to increase linearly with the amplitude of the microwave. One might also expect the nature of the defect, essentially normal conductor or insulator, to be crucial in defining both the density of free electrons in its interior and the boundary conditions of the Maxwell equations describing the system. Finally, one might hope to obtain in this way a quantitative understanding of the linear dependence of the residual resistance on the amplitude of the microwave. The similarities with the case of fluxon induced losses under study in the present work are worth noting. Both the residual resistance and the fluxon induced resistance depend linearly on the amplitude of the microwave, $R_{res} = R_{res}^0 + R_{res}^1 H_{rf}$ for the former and $R_{fl} = R_{fl}^0 + R_{fl}^1 H_{rf}$ for the latter (when talking about a single fluxon, H_{ext} should be omitted with the understanding that R_{fl}^0 and R_{fl}^1 are globally renormalised). Moreover the dependence of R_{fl}^1 on R_{fl}^0 is reminiscent of the dependence of R_{res}^1 on R_{res}^0 , more precisely the ratios R_{res}^1 / R_{res}^0 and R_{fl}^1 / R_{fl}^0 are of the same order of magnitude. This is illustrated in Figure 4 where the line drawn on the data of Figure 2 is superimposed on residual resistance data obtained for different films (Figure 5 of Reference [32]). In the fluxon pinning case the residual resistance induced by the pinning centres, which are expected to be “bubbles” having dimensions much smaller than the coherence length, has been shown to be very small [32], a consequence of the observation that the value of the residual resistance is nearly independent from the nature and concentration of noble gas in the film. Qualitatively, one might then expect the fluxon cores rather than the pinning centres to define the size of the reservoirs of unpaired electrons affected by the proximity effect and, to the extent that the amplitude of the microwave is not sufficient to unpin the fluxons, one might expect a linear dependence of R_{fl} on H_{rf} for the very same reason as in the case of R_{res} .

The very qualitative picture which has just been sketched should not be taken more seriously than it deserves. Its only ambition is to underline the major differences existing between the pinning configuration characteristic of the present work and that of the most commonly reported studies, and to bring to the attention of the reader the absence of relevant calculations in the literature, at least as far as we are aware. A last comment, of possible relevance to the present discussion, is to recall the intriguing enhancement of the H_{rf} dependence of fluxon induced losses in the presence of a strong electric field, well below the field emission threshold [37]. This enhancement has been shown to be responsible for the occasional R_{fl} kink briefly mentioned in the introduction.

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Table 1

A list of parameters characterising the properties of films A and B (or, for some of the parameters, averaged over a set of films prepared under the same conditions as films A and B). Most parameters are defined in the text. Δ is an approximate value of the gap [1], obtained from a fit of the BCS resistance to a form $R_{BCS}(T) \propto \exp(-\Delta/T) / T$, R_{BCS}^0 is $R_{BCS}(4.2K)$, λ_{rel} is the ratio of the penetration depth to its clean limit value, $a_{\perp}$ is the lattice parameter measured normally to the sample surface and σ_{in} is the average stress in the plane parallel to the surface.

Parameter	Film A (oxide-free Cu)	Film B (oxidised Cu)
R_{fl}^0 [nΩ/G]	55.6±0.8	3.3±0.2
R_{fl}^I [nΩ/G/mT]	4.51±0.13	0.91±0.01
Argon content [ppm]	286±43	435±70
T_c [K]	9.36±0.04	9.50±0.02
Δ [K]	18.1±0.8	18.7±0.7
R_{BCS}^0 [nΩ]	504±32	410±8
H_{thr} [G]	0.14±0.01	0.31±0.02
λ_{rel}	1.04±0.09	1.51±0.04
RRR	28.9±0.9	11.5±0.1
H_{c2} [T]	0.77±0.01	1.150±0.025
$a_{\perp}$ [Å]	3.3184±0.0006	3.3240±0.0010
σ_{in} [MPa]	-565 ± 78	-706 ± 56

Figure 1 - Dependence on $t^2 = (T/T_c)^2$ of the upper critical fields of samples A and B as measured at constant field (horizontal error bars) and at constant temperature (vertical error bars). The open squares are for measurements made with the film removed from the copper substrate. The dashed lines are the results of fits to the Gorter-Casimir form, $(1-t^2)/(1+t^2)$, the dotted lines to a form $(1-t^{1.7})^{1.1}$ observed to give a slightly lower χ^2 .

Figure 2 - Distribution of different films in the R_{fl}^l vs. R_{fl}^0 plane. Open symbols are for films of the A family and full symbols for films of the B family. Data points labelled with numbers are films grown using an argon-neon mixture, 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 3 - Distribution in the R_{fl}^I vs R_{fl}^0 plane of different films grown in argon atmosphere. The upper figure illustrates the separation in two families: a first one, in the upper right corner, including film A, and a second one, in the lower left corner, including film B. In the upper figure open symbols are for oxide-free substrates and full symbols for oxidised substrates. Details of a part of the B family are shown in the lower figure.

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 averages of films grown on hydroformed chemically polished copper (squares), on spun chemically polished copper (circles) and on spun electropolished copper (diamond). Also shown (triangle) is an average of data obtained with hydrogen loaded films. The line drawn across the data of Figure 2 is superimposed on the present figure and may be shifted arbitrarily along the diagonal, since it is only the ratio of the ordinate to the abscissa which is relevant to the comparison between R_{fl} and R_{res} .