Investigation of voltage steps of $U(I)$ curves in PbMo_6S_8 wires *

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We have investigated hot-drawn PbMo_6S_8 composite monofilament wires ($J_c = 4.6 \times 10^8 \text{ A/m}^2$ at $\mu_0 H = 10 \text{ T}$ and $T = 4.2 \text{ K}$) in particular steps that occur in the I – U transition curves. These steps have been measured in fields up to 9 T and in a temperature range between 2 K and 10 K. We have analysed them by considering two models, the hot spot model and the phase-slip model. Furthermore, a flux flow channel picture is developed, where the step is related to an avalanche of flux flow localised in a narrow channel. The observed field dependence of the step in a field applied transversally to the wire can be understood within this description. All these model considerations concur; the origin of the steps appears to lie in local defects with a μm length-scale. Just a few of these defects may already depress the overall critical current in a substantial way. Taking into account this situation, there exists an important potential for further improvements of the critical current in PbMo_6S_8 wires.

1. Introduction

The Chevrel-phase PbMo_6S_8 with its upper critical field [1] $\mu_0 H_{c2}$ of above 50 T is one of the most promising materials for future very high field applications of superconductors. Various groups are developing different wire technologies in order to improve the critical current density. During the last few years most groups have reached J_c -values at liquid helium temperature of 2×10^8 and $5 \times 10^8 \text{ A/m}^2$ at $\mu_0 H = 20 \text{ T}$ and 10 T, respectively [2–4].

The aim of this paper is to explore the present limitations of Chevrel-phase wires. In particular it is important to understand to what extent the present values represent an intrinsic limit or if J_c is still extrinsically limited, and to get insight into the nature of these limitations.

The measurements reported here were made on hot-drawn PbMo_6S_8 wires clad in a molybdenum (Mo) and stainless steel (SS) matrix. In comparison to copper these matrix materials have a rather high electrical resistance. This property, as well as an important interface resistance between the PbMo_6S_8 core and the Mo-SS-matrix [3,5] results in quite a poor thermal stability of the wire. Whilst in a practical wire this is of course not desired and has to be improved upon, this instability makes these wires particularly well suited to detect and locate weak spots limiting the critical current. These instabilities are leading in particular to steps in current voltage characteristics. As we will show, these voltage steps are related to local defects on the μm length-scale. Indeed, the high resistance matrix gives us a powerful tool to study the PbMo_6S_8 core in these wires.

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2. Sample preparation

The PbMo_6S_8 wires have been manufactured by a

powder metallurgy route. The prereacted powder was isostatically pressed, machined and passed into a molybdenum tube. This tube was fitted into a stainless steel tube which was, after outgasing, sealed under vacuum. The outer diameter of the whole billet was about 50 mm. The first reduction stage of the wire is made by hot extrusion at about 1200°C. Many subsequent drawing steps, between 900°C and 500°C (depending on the wire diameter), are necessary to reach the final wire diameter of 0.4 mm. Monofilamentary wires up to 1 km length have been manufactured.

3. Experimental results

In this section we give an overview of the various types of experimental results on critical current measurements. The measurements were performed on small coils, wound on a Al_2O_3 support with a diameter of 44 mm. The wire length was about 1 m and we used the $1 \mu\text{V}/\text{cm}$ criterion to determine the critical current. Up to 8 sections of the sample could be measured simultaneously by a computer controlled data acquisition system [3]. The superconducting cross section for different wires was between 1×10^{-8} and $4 \times 10^{-8} \text{ m}^2$.

The $U(I)$ transition curves from the superconducting to the normal state in our PbMo_6S_8 wires exhibit three different types of transitions as shown schematically in fig. 1. The round transition is observed for current densities (J_c) typically below $2 \times 10^8 \text{ A/m}^2$. The transition curve showing voltage jumps (steps) is observed for J_c values between 2×10^8 and $4 \times 10^8 \text{ A/m}^2$. The thermal run away, where a propagating normal zone drives the entire sample into the resistive state, is observed for J_c values higher than $4 \times 10^8 \text{ A/m}^2$.

Two experimental curves measured at $T=9 \text{ K}$ are represented in fig. 2 where the 1 T curve is a typical

Fig. 1. The superconducting $U(I)$ transition: (a) round, (b) step and (c) thermal run away.

Fig. 2. $U(I)$ transition curves of a PbMo_6S_8 wire at $T=9 \text{ K}$ in transverse magnetic field of 0.5 T and 1 T. The numbers refer to the differential resistance of the linear part of each step.

case for the round transition. The round transition is the usual transition curve for composite wires. The curvature is directly related to the J_c -distribution in the wire and there are various ways to extract it. The mechanism of the round transition is well understood (see for instance Warnes [6], Plummer and Evetts [7] and Hampshire and Jones [8]) and, as it is observed, as in this particular case, only in samples with low J_c , we will focus our discussion on the other types of the superconducting transition.

The curve at 0.5 T shows several steps very similar to those observed by Akhmetov and Baev [9] in Nb_3Sn composite conductors and by Meyer [10] in tin whiskers. It may be noted that measurements with decreasing currents lead to a hysteresis which is characteristic for these transitions.

At higher current, for instance at lower fields or temperatures, an abrupt thermal run away of the wire is observed. In this case the heat balance becomes unstable and the temperature rises. With an oscilloscope we could observe [3] the rise of the resistance due to a normal conducting front travelling with approximately constant velocity through the whole sample. The complete sample was turned into the normal state within one second. The origin of this behavior is related to a defect in the wire where the local critical current is exceeded. This defect may cause a step or the thermal run away behavior, depending on temperature, field and the cooling rate. The latter could be shown in an experiment [3] where a sample that initially showed a thermal run

away for a sample current I_T , was soldered into a copper-profile. This is a configuration in which the heat exchange to the helium bath is strongly enhanced. The transition curve then showed a voltage step at the same sample current I_T , thus demonstrating that the thermal run away is related to a voltage step. We shall therefore concentrate on the study of the voltage steps and their physical origin.

4. Steps

A representative example for a single step is shown in fig. 3. Similar curves have been observed in many other samples. They can be characterized as follows: as the current in the sample is increased, zero voltage is observed up to a value I_{st} . At $I = I_{st}$ a voltage of typically 100 μV appears instantaneously. After the development of the step, the voltage increases linearly, the extrapolation to zero voltage intersects the current axis far from the origin at a value $I \geq I_{st}/2$. From this fact we learn that the voltage cannot simply be associated with a normal zone of constant size in the wire. When the current in the wire is decreased, a pronounced hysteresis is observed: first the voltage follows the same straight line down to the current I_{st} . At this value the voltage does not drop to zero. Only when the current is decreased down to $I_r < I_{st}$ the voltage falls to zero. This collapse is often preceded by a slight downward curvature of the $U(I)$ characteristic. The reproducibility of the $U(I)$ curve between two measurements is perfect if the sample is not warmed up to room temperature in between. Thermal cycles sometimes lead to a decrease of I_{st} .

It has been shown [3] by bringing the voltage taps

Fig. 3. Typical steps in PbMo_6S_8 wires at $T = 4.2$ K. The field is orientated perpendicular to its axis.

closer, that the step voltage is generated in less than 1 mm of the sample length. This indicates that the step is developed at a weak point of the sample which we will refer to as a defect. For the case when more than one step is observed each step is associated to a distinct defect at different sites in the wire.

The most frequently observed defects in these wires are cracks. The study of micrographs reveals different kinds of cracks in the PbMo_6S_8 core. Cracks oriented perpendicular to the wire axis are attributed to the relaxation of thermal stress due to a mismatch of thermal expansion between the matrix and the superconducting core [11]. There are also cracks parallel to the wire axis which are probably an artefact of the preparation of the micrograph (release of stress when the wire is opened). Wires with steps in the $U(I)$ characteristic do not systematically show visible cracks in the limit of resolution of our SEM. Of course any other local reduction of the effective cross section, for example bad grain boundaries, must also be considered as defect candidates.

Figure 4 shows the field dependence of a step. For clarity the hysteresis is omitted and only curves with increasing currents are reported. The field is orientated perpendicular to the wire. The step height decreases when the field is enhanced and the slope of the linear part increases for fields higher than 3 T (inset (a)) and may alternatively be fitted to an $dU/dI = a + bH$ behavior or to a $dU/dI = a\sqrt{H}$ behavior as will be discussed. The low field behavior can be attributed to temperature variations related to high measuring currents. The step current $I_{st}(H)$, is a de-

Fig. 4. Field dependence of the step for increasing current. The field is orientated perpendicular to the wire.

creasing function of the field and is shown in inset (b).

Figure 5 shows the temperature dependence of a step. Again, hysteresis is omitted. The three steps which line up one to each other have been measured in liquid He at 4.2 K, 3.2 K and 2.6 K. We assume that the deviation from this continuous behavior is due to a change in the heat transfer conditions. Indeed below the λ -point (2.1 K curve) the heat transfer is strongly enhanced. On the high temperature side (5 K, 6 K curves, measurements are made in helium gas flow) the heat transfer to the He gas is poor and a displacement of the curves in the opposite direction is observed. As can be seen from the inset, the slope of these steps also depends on these modifications in the cooling of the sample. We interpret these facts as evidence that thermal effects play a role in the development of the step.

5. Discussion

The above described results are very similar to the $U(I)$ curves observed in Nb_3Sn wires [9], tin whiskers [10] and in microbridge samples [12–14]. Although the precise geometry in our case is not entirely known, and probably different, we will discuss our results in the light of the two models developed by Skocpol et al. for their microbridge samples, made of Sn films. There is however some difficulty in using these models to interpret our data. This is due to the existence of the normal conducting matrix of the

wires which acts as a shunt parallel to the defect. As we have shown by a model calculation [3,5], an interface resistance between the superconducting filament and the matrix leads to a current-transfer length of several 100 μm . This implies that the current is conserved in the superconducting filament when the length of the resistive defect in the $\text{Pb-Mo}_6\text{S}_8$ core is small in comparison to this length. Under this condition the influence of the electrical conduction of the matrix can be neglected and we can compare our measurement directly to model considerations which are dealing with the superconductor only.

5.1. Hot spot model

The idea underlying the hot spot model can be expressed as follows: at a weak point of the sample, the critical current (depending on field and temperature) is exceeded locally and power dissipation occurs. If the thermal contact to the He-bath is poor, the temperature rises above the critical temperature and a normal domain develops rapidly. The abrupt rise of the sample resistance is then visible as a step in the $U(I)$ characteristic. The final size of the hot spot depends on the thermal balance between heat generation due to Joule effect and heat evacuation by conduction to the helium bath. It turns out that this equilibrium size of the hot spot is not constant, i.e. it rises when the current is further enhanced and it shrinks when the current is lowered. This explains, why the slope does not follow Ohm's law which would extrapolate to the origin. It has been shown [3], that the complete picture as described (step, linear slope and hysteresis) emerges if a crack-like geometry of the microbridge sample is assumed. We take this as an indication that our defects are related to a reduction of the active PbMo_6S_8 cross section.

An original explanation of the steps, related to the hot spot picture has been given by Akhmetov and Baev [9]. They observed multiple voltage steps in stabilised Nb_3Sn wires with a high interface resistivity, similar to our 0.5 T curve in fig. 2. All measurements reported have been realised on the total length of 0.5 m of their samples. The generation of the first step is related to the generation of a normal zone of finite size. In their description the second step is not the repetition of the creation of another

Fig. 5. Temperature dependence of the step for increasing current. The field is orientated perpendicular to the wire.

distinct normal conducting zone but it is related to the existence of the first normal zone: in the middle of the normal zone a superconducting region is created dividing the normal zone into two distinct domains. All the following steps are the result of the subdivision of normal zones. Akhmetov reports also the experimental differential resistance dU/dI of the slope of the step. His curve follows a linear behavior, $dU/dI = a + bH$ also at low fields.

The creation of multiple normal zones which is related to the existence of one of the preceding normal zones is of course in contradiction to our experimental findings where each step is related to a distinct location (distant by several cm, sometimes several tenths of a cm) in the sample. Further, our time resolved observations are indicating a continuous increase of the voltage when we are in the thermal run away regime and not a step like increase as is observed by Akhmetov and Baev. This also indicates that their description is not valuable for our observations.

A thermal analysis [3] based on Fourier's heat equation leads to a minimum size for the existence of the normal conducting hot spots in PMS. We evaluated this size to be of the order of $10\text{ }\mu\text{m}$ in PbMo_6S_8 wire samples. The corresponding step height is calculated to be of the order of several mV which is much more than we usually observe. For this reason we do not believe that the hot spot picture is the appropriate description for our steps.

For a complete description of the hot spot problem we refer to an extensive review that has been made by Gurevich and Mints [15].

5.2. Phase-slip model

The idea underlying the phase-slip model can be expressed as follows: before the critical depairing velocity of the Cooper pairs is reached, the superconductor enters a dissipative phase-slip mode. This mode was first considered by Langer and Ambegaokar [16]. Skocpol et al. [12] have established a phase-slip model for tin microbridges. This model explains the step by the activation of a phase-slip center at a weak point of the sample (defect). When the characteristic current I_{st} of the phase-slip centre is exceeded, the total sample current can no longer be carried by the Cooper pairs. Actually a dissipa-

tive, normal current must make up the difference. This means that before entering the phase-slip center some of the Cooper pairs are broken up into quasiparticles which cross the phase-slip center and then recombine to Cooper pairs on the other side. The characteristic length over which normal electrons participate to the current transport is the quasiparticle diffusion length. For the case of PbMo_6S_8 this length is estimated [3] to be of the order of one tenth of a μm .

The theoretical transition curves for tin microbridges, based on the phase-slip mechanism, are shown in fig. 6 of ref. [14]. As can be seen there, all the features of the step that have been observed in our PbMo_6S_8 samples are qualitatively reproduced in curves E and F if heating effects are considered. As for the experimental curves the step-voltage is of the order of $100\text{ }\mu\text{V}$. The measuring current in the microbridge samples is much smaller than the current in our sample. Hence the current density turns out to be about the same in the two cases.

The calculations leading to their curves had been carried out for a quasi-one-dimensional superconductor which means that the transverse dimension of the superconductor d is small in comparison with the coherence length ξ . It is clear that this condition cannot be exactly fulfilled in our wires, first because of the large transverse dimensions of the superconductor ($100\text{ }\mu\text{m}$ nominal diameter) and second because of the very small coherence length of about $\xi_{GL} = 28\text{ }\text{\AA}$ in PbMo_6S_8 . Of course, it is possible that the weak spot corresponds to a very narrow connection between two grains approaching the conditions for this phase-slip analysis; however, to be fully valid for our samples, the phase-slip model should be generalized to the three-dimensional case.

Another point is that most of our measurements are made in a transverse magnetic field higher than the lower critical field of $B_{c1} = 5\text{ mT}$. Under these conditions a type-II superconductor is in the vortex state and it is not clear if the phase-slip picture would be a good description of the evolution of the phase in our samples. For this reason we will now present an approach based on the activation of flux flow in a narrow channel located on the defect.

5.3. Flux flow channel (FFC)

The idea to use the flux flow picture to describe the steps is motivated by the hot spot model. At the defect position indicated in fig. 6, J_c is reached for a transport current I which can still flow without dissipation in the other parts of the wire. This means that only the flux lines in a small channel at the defect position begin to move and dissipation occurs locally. If this dissipation exceeds the power threshold for the hot spot generation, a normal hot spot is created and the flux lines disappear. If, on the other hand, dissipation remains small, superconductivity is not destroyed and the flux continues to flow. In this case we should observe a $U(I)$ curve according to $U = R_f (I - I_c)$ [17]. Under these conditions a step can only be observed if the flux lines at the defect site are retained until the current $I_{st} > I_c$ is reached.

The phenomenon is related to a similar mechanism as in magnetisation experiments when flux jumping occurs. In such experiments the magnetic field gradient applies a force on the flux line lattice (FLL). When the critical force is reached flux jumping takes place, the magnetic field gradient is released and no new flux jumping occurs if the field is not further enhanced.

In our case, the interaction between the flux lines inside and outside the FFC could explain the stabilisation of the flux lines until the step current is reached. At this current the flux motion sets in abruptly, resulting in a voltage step. An important difference with magnetisation experiments is that the driving force, which is here the transport current, is not released when the flux is moving. This has the

Fig. 6. Local onset of flux flow at a defect location.

consequence that the flux motion will not stop and we continue to observe the step voltage. When the current is further enhanced, the voltage follows the usual flux flow behavior leading in a most natural way to the extrapolation of $U(I)$ to non zero currents.

It is interesting to notice that the flux flow picture also conserves aspects from the phase-slip model: in both cases superconductivity is not destroyed. Part of the current continues to flow without dissipation and the phase correlation is conserved. In the phase-slip model each phase-slip is associated with a phase variation of 2π . In the flux flow picture the flux lines are moving perpendicularly to the current direction. Each flux line crossing the superconductor contributes (as a phase-slip) to a phase variation of $\Delta\phi = 2\pi$. We can use this fact to make an estimate of the slope of the linear part of the step. The voltage drop is related to the phase variation by the Josephson relation:

$$U = \frac{h}{2e} \frac{\partial \phi}{\partial t}. \quad (1)$$

The phase evolution can be found, by computing the rate dn/dt at which the flux lines cross the filament:

$$\frac{\partial \phi}{\partial t} = 2\pi \frac{dn}{dt} = 2\pi \rho_v v_f d, \quad (2)$$

where ρ_v is the field-dependent flux line density and d the defect length over which flux flow occurs (fig. 6). The flux flow velocity v_f is found by equating the driving Lorentz force to the viscous force ($J - J_c$) $\phi_0 = \eta v_f$. Here ϕ_0 is the flux quantum and η is the viscosity of the flux motion which can be approximated by the viscosity of Bardeen and Stephen [18] $\eta = (\phi_0 B_{c2}) / \rho_N$. Here ρ_N is $(2-3 \mu\Omega \text{ m})$ [19] the normal state resistivity of PbMo_6S_8 .

We can estimate the field dependence of the $U(I)$ curve by combining these equations and taking the derivative dU/dI :

$$\frac{dU}{dI} = \frac{d}{A_{\text{eff}}} \rho_N \frac{B}{B_{c2}}. \quad (3)$$

We have used $J = I/A_{\text{eff}}$ for the current density at the defect in the resistive state, where A_{eff} is the effective cross-section for the current (fig. 6). The right-hand side is exactly the flux flow resistance of a sample with a cross-section A_{eff} and a length d .

The experimental data are shown in inset (a) of fig. 4, which can be described by a $dU/dI = a + bH$ behavior for fields higher than 3 T. We can estimate the geometrical extension of the FFC by a fit of the experimental values dU/dI to eq. (3). When using an effective B_{c2} value of 35 T–60 T we find that $d/A_{\text{eff}} \approx 35 \text{ m}^{-1}$ is almost independent of temperature in the range of $T = 2 \text{ K} - 5 \text{ K}$. An estimate for the maximum value of d can be found if we use the fact that A_{eff} is smaller than the superconducting filament section A_{PMS} . We then find a d value in the range of $0.5 - 1 \text{ } \mu\text{m}$. At a magnetic field $\mu_0 H = 9 \text{ T}$, the flux line density is $4 \times 10^{15} \text{ m}^{-2}$. This leads to the result that less than 50 rows of flux lines can find a place in the FFC. Our presumption that it is the interaction of flux lines inside and outside the FFC which is responsible for the voltage-step behavior becomes more plausible if the number of flux lines that have to be retained is small.

We again find that the step voltage is generated within a very short length of the sample.

5.4. Single row FFC

The linear fit of the field dependence of dU/dI for higher fields, see inset (a) in fig. 4, does not extrapolate to the origin. This offset is at present not understood and in this context we have so far no explanation for this behavior. Nevertheless, the high-field data ($\mu_0 H > 3 \text{ T}$), can also be nicely fitted to a $\sqrt{H}$ behavior without offset as is shown in fig. 7. The deviation from the $\sqrt{H}$ behavior can then be explained just as for the linear case by heating effects which become important for high measuring currents at low fields. A $\sqrt{H}$ behavior can be understood quite easily: we just have to assume that the FFC is so narrow that it barely holds a single row of flux lines. In this case, only the linear density of flux lines, which is proportional to $\sqrt{H}$, enters eq. (2). If we then repeat the calculation for dU/dI , we find the following relationship:

$$\frac{dU}{dI} = \frac{\rho_N}{A_{\text{eff}}} \frac{\sqrt{\phi_0 B}}{B_{c2}}. \quad (4)$$

This result no longer depends on the width of the FFC. In fact, the length of dissipation d is of the order of the FLL-constant, which is even smaller than for the previous case. The fit to our experimental data

Fig. 7. $(dU/dI)^2$ field dependence of the linear part of the step from fig. 4. The $\sqrt{H}$ dependence, shown by the dotted line, is followed for fields ($\mu_0 H$) smaller than 3 T.

allows us to find the estimate for $A_{\text{eff}} \approx 20 \text{ } \mu\text{m}^2$ which is now the only free parameter. This result leads to a value for the local critical current density of more than 10^{10} A/m^2 . A confirmation of the $\sqrt{H}$ behavior would therefore be a very important result.

A $\sqrt{H}$ dependence has recently been proposed by Pan et al. [20] for flux flow in low angle grain boundaries in YBaCuO. The $U(I)$ curves discussed there are showing variations of the slope but there are no voltage steps. These curves are explained as a consequence of the onset of flux flow in an increasing number of one-dimensional easy slip channels. In our case the $U(I)$ curves are the result of the flux line motion at a defect location. Of course, grain boundaries are present in a great number in our PbMo_6S_8 which was fabricated by the powder metallurgical route. It is therefore very plausible that one or even several grain boundaries are activated when the local critical current at a defect site is exceeded.

6. Conclusions

We have investigated the superconducting transitions of PbMo_6S_8 wires. These wires have a stainless steel Mo matrix, where the Mo acts as a diffusion barrier for the PbMo_6S_8 . The poor thermal and electrical conductivity of the matrix allows us to uncover the presence of weak spots in the wire producing steps or even thermal runaway behavior in the $U(I)$ characteristics.

The hot spot model and the phase-slip model both can in principle provide an understanding of voltage steps. However, for PbMo_6S_8 the hot spot model leads to voltage steps of a height of several mV which is much more than we observe in our experiments. Neither model so far can easily explain the field dependence of the steps in a transverse field. The flux flow channel (FFC) picture developed above can provide an explanation of local motion of flux lines in grain boundaries at defect locations. The FFC picture gives a qualitative understanding of the steps and can describe a H or a $\sqrt{H}$ dependence of the linear part of the step. Despite this uncertainty in the interpretation of the flux channel model, this is the only description which explains the field dependence of the steps. If the $\sqrt{H}$ dependence turns out to be correct, the voltage steps can be understood by a single-row FFC. In this case the local critical current density is found to be higher than 10^{10} A/m^2 at a field of 9 T.

The experimental findings and the various theoretical considerations are clearly leading to the conclusion that local defects of sizes in the μm -range and sub- μm -range are determining the critical current in these wires. The defects are most likely due to microcracks in the SC filament, or related to a local variation of the quality of grain boundaries. The fact that in a sample of length 1 m, less than ten voltage steps are observed indicates that there are only a few defects depressing the observed I_c value. Between these defects I_c is certainly higher, maybe much higher. The J_c is therefore still extrinsically limited and the suppression of these defects should lead to an improvement of the performances of these wires.

We finally note that various groups have all reached J_c values in PbMo_6S_8 wires of 2×10^8 and $5 \times 10^8 \text{ A/m}^2$ at $\mu_0 H = 20 \text{ T}$ and 10 T respectively. It might be possible that all groups are limited by the same local defects. The difference is that in a low resistance matrix the voltage steps associated with the defects cannot be observed. A model calculation [3,5] has shown that the high resistance Mo-stainless steel matrix is necessary to observe steps and to detect the presence of defects in the wire.

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