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High Field determination of superconducting fluctuations in high-\(T_c\) cuprates

F. Rullier-Albenque\(^1\), H. Alloul\(^2\), D. Colson\(^1\) and A. Forget \(^1\)

\(^1\) Service de Physique de l’Etat Condensé, Orme des Merisiers, CEA Saclay (CNRS URA 2464), 91191 Gif sur Yvette cedex, France

\(^2\) Laboratoire de Physique des Solides, UMR CNRS 8502, Université Paris Sud, 91405 Orsay, France

E-mail: florence.albenque-rullier@cea.fr

Abstract. Large pulsed magnetic fields up to 60 Tesla are used to suppress the contribution of superconducting fluctuations (SCF) to the ab-plane conductivity above \(T_c\) in a series of \(\text{YBa}_2\text{Cu}_3\text{O}_{6+x}\) single crystals. The fluctuation conductivity is found to vanish nearly exponentially with temperature, allowing us to determine precisely the field \(H'_c(T)\) and the temperature \(T'_c\) above which the SCFs are fully suppressed. \(T'_c\) is always found much smaller than the pseudogap temperature. A careful investigation near optimal doping shows that \(T'_c\) is higher than the pseudogap \(T^*\), which indicates that pseudogap cannot be assigned to preformed pairs. For nearly optimally doped samples, the fluctuation conductivity can be accounted for by gaussian fluctuations following the Ginzburg-Landau scheme. A phase fluctuation contribution might be invoked for the most underdoped samples in a 

1. Introduction

One of the most puzzling feature of the high-\(T_c\) cuprates is the existence of the so-called pseudogap phase in the underdoped region of their phase diagram. After the first evidence of an anomalous drop of the spin susceptibility detected by NMR experiments in underdoped YBCO well above \(T_c\) \(^1\), a lot of unusual properties have been observed in the pseudogap phase \(^2\). However its exact relationship with superconductivity is still highly debated. In fact there does not exist up to now a unique representation for the pseudogap line \(T^*\) as illustrated in Fig\(^1\). Either \(T^*\) is found to merge with the superconducting dome in the overdoped part of the phase diagram, or to cross it near optimal doping. In the first case, it has been proposed that the pseudogap could be ascribed to the formation of superconducting pairs with strong phase fluctuations \(^3\). This scenario has been supported by the observation of a large Nernst effect and of diamagnetism above \(T_c\), which delineates another line \(T'_c\) below which strong superconducting fluctuations and/or vortices persist in the normal state \(^4\). In the second approach, the pseudogap and the superconducting phases arise from different, even competing, underlying mechanisms and are associated with different energy scales \(^5\).
While superconducting fluctuations (SCF) are expected to be especially large in these anisotropic compounds with low superfluid density, there is not a clear consensus about the temperature range above \( T_c \) in which SCFs survive. In this paper, we will present our results on superconducting fluctuations for a series of YBa\(_2\)Cu\(_3\)O\(_{6+x}\) single crystals from underdoping to slight overdoping. We have used an original method based on the measurements of the magnetoresistance in high pulsed magnetic fields. We have initiated this method in ref.\([6]\) in underdoped compounds, and have done complementary measurements for various O contents \([7]\). In this later paper we gave an extensive report on the data and their analysis. Here we present a simplified comprehensive summary of our main results and discuss them in the context of recent experimental results reported by others at this conference.

The principle of our study is to use high magnetic fields to determine the normal state resistivity and to extract with high accuracy the superconducting fluctuations (SCF) contributions to the conductivity and their dependences as a function of temperature and magnetic field (section 2). We are thus able to determine the threshold values of the magnetic field \( H'_c \) and temperature \( T'_c \) above which the normal state is completely restored. In the same set of transport data, we could compare the values of \( T'_c \) and of the pseudogap temperature \( T^* \) as a function of doping \([8]\) (sec.3). We have added in the present paper another comparison of these two temperature scales by using previous NMR data to determine \( T^* \) \([1]\). We will also show how our results can be analysed in the framework of the Ginzburg-Landau approach, making it possible to extract microscopic parameters of the superconducting state such as the zero-temperature coherence length (sec.4-5). The effect of disorder introduced by electron irradiation at low temperature will be also presented (sec.6).

2. Experimental

Details on the experimental conditions concerning the different single crystals and the high-field experiments as well as the method used to extract the SCF contribution to the conductivity are given in ref.\([7]\). Four different single crystals of YBa\(_2\)Cu\(_3\)O\(_{6+x}\) have been studied. They are labelled with respect to their critical temperatures measured at the mid-point of the resistive transition: two underdoped samples UD57 and UD85, an optimally doped sample OPT93.6 and a slightly overdoped one OD92.5, corresponding to oxygen contents of approximately 6.54, 6.8, 6.91 and 6.95 respectively. Some of these samples have been irradiated by electrons at low \( T \), which allows us to introduce a well controlled concentration of defects in the CuO\(_2\) planes \([9]\).

The transverse MR of the different samples have been measured in a pulse field magnet up to 60T at the LNCMI in Toulouse. An example of the transverse MR curves measured on the OPT93.6 sample is illustrated in fig.2 for \( T \) ranging from above \( T_c \) to 150K. In the normal state well above \( T_c \), it was shown that the transverse MR increases as \( H^2 \) both in optimally doped and in underdoped YBCO \([10]\). This is indeed what is found also here for \( H \) up to 60T and for \( T \gtrsim 140K \) (see inset of fig.2). At lower \( T \), some downward departure from this \( H^2 \) behavior is observed for low values of \( H \) which we attribute to the destruction of SCFs by the magnetic...
field. The normal state behavior is only restored above a threshold field \( H'_c(T) \) which increases with decreasing temperatures. In ref.\[7\], we have shown that the magnetoresistance coefficients measured at low \( T \) above \( H'_c(T) \) are in continuity with those measured at higher temperatures and low field, which is a strong indication that the effect of the field is to restore the normal state.

This experimental approach allows us to single out the normal state properties and determine the SCF contributions to the transport. In particular, the extrapolation down to \( H = 0 \) of the \( H^2 \) normal state MR above \( H'_c(T) \) gives us the value of the normal state resistivity \( \rho_n(T) \). The way to extract the fluctuating conductivity and its dependence with temperature \( \Delta \sigma_{SF}(T,0) \) and magnetic field \( \Delta \sigma_{SF}(T,H) \) is explained in details in ref.\[7\].

3. Onset of superconducting fluctuations and pseudogap

The \( T \) dependences of the zero-field superconducting conductivities \( \Delta \sigma_{SF}(T,0) \) are reported in Fig.3 for the OPT93.6 and UD57 samples and compared to the off-diagonal Peltier conductivity deduced from the Nernst measurements on the same samples \[11\]. We observe that \( T'_c \) is always found larger that the onset of Nernst signal \( T'_{\nu} \). This might come from the difficulty to choose a good criterion to determine \( T_{\nu} \) as the minimum in \( \alpha_{xy}/B \) may hide the real onset of SCFs. On the contrary, in Fig.4 where \( \Delta \sigma_{SF}(T,0) \) are plotted in a semi-logarithmic scale, one can note that this quantity vanishes very fast. This allows us to define a precise criterion to determine the onset temperature \( T'_c \), corresponding here to \( \Delta \sigma_{SF}(T,0) \equiv 1 \times 10^{-3} (\Omega m)^{-1} \). Of course decreasing or improving the sensitivity for SCF detection will result in decrease or increase of \( T'_c \), which might explain the different temperature ranges of SCFs derived from different experimental probes. If we were able to improve our sensitivity by an order of magnitude, the values of \( T'_c \) could only be increased by \( \sim 15 \text{K} \) and would correspond to an extremely low SCF contributions to the conductivity, about four orders of magnitude lower than at measured at \( T_c \).

In the case of the optimally doped compound, our value of \( T'_c \) is in very good agreement with the onset temperature determined by magnetic susceptibility \[12\]. However these values are found larger than those determined by microwave ac conductivity measurements \[13\]. This can be likely ascribed to the fact that a field of 16T is assumed to be sufficient to suppress all superconductivity above the zero-field \( T_c \) in this latter work, which is clearly in contradiction with our results. On the other hand, recent terahertz conductivity measurements in LSCO show that the signatures of the fluctuations only persist in a very narrow range, at most 16K above
This has to be contrasted with determinations from Nernst measurements [4] or high field magnetoresistance measurements comparable to ours [15] which give much larger onset temperatures. This clearly indicates that these different types of experiments do not detect the SCFs with a similar sensitivity as ours, or that they probe different types of SCFs (namely phase versus amplitude).

Independently of these remarks, all the recent experimental data now point to an onset temperature for superconducting fluctuations well below the pseudogap temperature $T_c$. One can also observe in Fig.5 that $T'_c$ is only slightly dependent on hole doping, increasing from $\sim 120$K to $\sim 140$K from the UD57 sample to the optimally doped one OPT93.6. This is very similar to what has been found from Nernst or magnetization experiments in Bi2212 [16]. However this strongly contrasts with the pseudogap temperature $T^*$ which decreases with increasing doping.

In order to compare the extension of the SCFs with respect to the opening of the pseudogap, the drop of the electronic susceptibility as measured by the Y NMR Knight shift [17] is reported in Fig.6 together with the fluctuation conductivity for the UD57 sample. These data clearly evidence that the electronic states lost by the opening of the pseudogap at $T^*$ are not redistributed into the formation of preformed pairs in a precursor superconducting state as nearly half of the susceptibility has already been suppressed at $T'_c$. This strongly indicates that these two states are not related. The situation is more delicate for the optimally doped sample for which $T^*$ becomes comparable to $T'_c$. In this case, a careful examination of the resistivity data allowed us to determine both the onset of SCFs and the pseudogap temperature with the same experimental sensitivity criterion [8]. The fact that the $T'_c$ line crosses the pseudogap line near optimal doping, as reported in Fig.5, unambiguously proves that the pseudogap phase cannot be a precursor state for superconductivity.

4. Quantitative analysis of the paraconductivity in the Ginzburg-Landau approach

The variations of $\Delta \sigma_{SF}(T)$ are reported versus $\epsilon = \ln(T/T_c)$ in Fig.7 for the four hole dopings studied. Except for the UD57 sample, it is striking to see that the experimental
The values of $T'_c$ (■) and $T^*$ (●) are plotted versus the hole doping for the four samples studied (from ref.[7]). The solid line indicates the superconducting dome. Contrary to $T'_c$ that is rather insensitive to hole doping, $T^*$ is found to decrease with increasing doping and intersects the $T'_c$ line near optimal doping [8].

Figure 6. The fluctuation conductivity (●, left scale) is compared to the Y NMR Knight shift [1], [17] (■, right scale) for the UD57 sample. It is remarkable to see that nearly half of the susceptibility has already been lost at the onset of SCFs.

Data collapse on a single curve. For $\epsilon \lesssim 0.1$ these results can be well accounted for by gaussian fluctuations within the Ginzburg-Landau (GL) theory [18]. In this approach the excess fluctuating conductivity, called here paraconductivity, is related to the temperature dependence of $\xi(T)$, the superconducting correlation length of the short-lived Cooper pairs, which is expected to diverge with decreasing temperature as:

$$\xi(T) = \xi(0)/\sqrt{\epsilon},$$

where $\xi(0)$ is the zero-temperature coherence length and $\epsilon = \ln(T/T_c) \simeq (T - T_c)/T_c$ for $T \gtrsim T_c$. More generally, the temperature dependence of $\Delta \sigma_{SF}(T)$ is given by the Lawrence-Doniach (LD)
expression which takes into account the layered structure of the high-$T_c$ cuprates [19]:

$$\Delta\sigma^{LD}(T) = \frac{e^2}{16\hbar s} \frac{1}{\epsilon \sqrt{1 + 2\alpha}}. \quad (2)$$

where the coupling parameter $\alpha = 2(\xi_c(T)/s)^2$ with $\xi_c(T) = \xi_c(0)/\sqrt{\epsilon}$. Sufficiently far from $T_c$, one expects $\xi_c(T) \ll s$ and Eq.2 reduces to the well-known 2D Aslamazov-Larkin expression:

$$\Delta\sigma^{AL}(T) = \frac{e^2}{16\hbar s} \epsilon^{-1} = \frac{e^2}{16\hbar s} \xi^2(0). \quad (3)$$

The only parameters in this expression are the value of the interlayer distance $s$ and the value taken for $T_c$ which can have a huge incidence on the shape of the curve especially for $(T - T_c)/T_c < 0.01$. It can be seen in Fig.7 that the data are reasonably fitted by the LD expression (Eq.2) in the small temperature range $0.03 \leq \epsilon \leq 0.1$ if one takes $\xi_c(0) \simeq 0.9\AA$. We have assumed here, as usually done, that the CuO$_2$ bilayer constitutes the basic two-dimensional unit, and $s$ is then taken as the unit-cell size in the $c$ direction: $s = 11.7\AA$.

For the UD57 sample, $\Delta\sigma_{SF}(T)$ is found to be much larger (about a factor four at $\epsilon = 0.05$) than for the other doping contents. Quite surprisingly, it is possible to recover a good matching with these latter data by assuming an effective mean field temperature $T_{0}\tilde{\epsilon}$ different from the actual $T_c$. This is illustrated by the empty symbols in fig.7 using $T_{0\tilde{\epsilon}} = 72K$. This points to an additional origin of SCFs below $T_{0\tilde{\epsilon}}$ which might be ascribed to phase fluctuations of the order parameter. This is discussed in more details in ref.[7].

For all the samples, one can see in fig.7 that $\Delta\sigma_{SF}(T)$ vanishes very rapidly for $\epsilon \gtrsim 0.1$. This behaviour which has been noticed previously in many studies is particularly well defined here given the method used to extract the fluctuating conductivity. An extension of the AL theory including short wave length fluctuations [20] has been invoked to explain the steeper decrease of $\Delta\sigma_{SF}(T)$. This has also been treated by inserting a cutoff phenomenologically, implying that the density of fluctuating pairs vanishes at $T_c$ as detailed in ref.[7].

5. Field variation of the SCF conductivity: $H'_c$ and upper critical field $H_{c2}$

From the data reported in Fig.2, we can also extract the magnetic field $H'_c(T)$ above which the MR recovers a $H^2$ dependence, which we take as the sign that the normal state is completely restored. As $T$ decreases, it becomes difficult to ascertain that the normal state is fully reached when $H'_c(T)$ becomes comparable to the highest available field. This makes it difficult to precisely deduce values of $H'_c(T)$ larger than 45T.

The evolution of $H'_c(T)$ are plotted in fig.8 for the four samples. One can see that $H'_c(T)$ increases rapidly with decreasing $T$ and displays a linear variation near $T'_c$. We have tentatively tried to estimate a low $T$ extrapolation of $H'_c$ using a parabolic $T$ variation as applied for the critical field of classical superconductors:

$$H'_c(T) = H'_c(0)[1 - (T/T'_c)^2]. \quad (4)$$

The fitting curves are displayed as dashed lines in Fig.8 and show that $H'_c(0)$ increases with hole doping and reaches a value as high as $\sim 150$ Tesla at optimal doping.

A precise analysis of the fluctuation magnetoconductivity $\Delta\sigma_H(T,H)$ is a valuable tool to extract different microscopic parameters of high-$T_c$ cuprates, such as the value of $H_{c2}(0)$ not directly accessible from experiments. $\Delta\sigma_H(T,H)$ can be written out as:

$$\Delta\sigma_H(T,H) = \Delta\sigma(T,H) - \Delta\sigma_n(T,H) = \Delta\sigma_{SF}(T,H) - \Delta\sigma_{SF}(T,0). \quad (5)$$
It has been very often assumed that the second term of the first equation can be neglected as being only weakly dependent on magnetic fields. However, our study clearly shows that this is not the case (see for instance the data displayed in fig.2). Thus our method provides here a correct determination of $\Delta \sigma_H(T,H)$.

In the GL approach, the evolution of the fluctuation magnetoconductivity with $H$ comes from the pair-breaking effect which leads to a $T_c$ suppression. In the case of interest, the major contribution results from the AL process, and more particularly from the interaction of the field with the carrier orbital (ALO) degrees of freedom. The detailed analysis and discussion of the fluctuation magnetoconductivity are reported in ref.[7]. The important point here is that the analysis of the fluctuation magnetoconductivity can give a direct determination of the coherence length, and then of $H_{c2}(0)$, as the same coherence length governs the fluctuating and the superconducting regimes. Thus a mirror field $H^*(T)$ of the upper critical field $H_{c2}(T)$ can be determined above $T_c$ in the GL approach [18]. Fig.9 shows the data for the UD85 sample together with the fits using the ALO expression with $H_{c2}(0) = 125(5)$T being the only adjustable parameter. We checked that, as predicted by the theory, the fits are valid as long as $H \lesssim H^*(T) = \epsilon H_{c2}(0)$.

Let us mention here that the analysis of the magnetoconductivity in terms of the ALO expression has to be restricted to the temperature range where the fluctuating conductivity can be also well described within the GL framework. For instance for the UD-85 sample, it is not possible to fit the $\Delta \sigma_H(H,T)$ curves with the same value of $H_{c2}$ for $T > 99$K ($\epsilon > 0.16$) above which $\Delta \sigma_{SF}(T)$ starts to deviate significantly from the LD expression (see fig.7). This would give values of $H_{c2}$ correspondingly smaller as the temperature is higher. Moreover, in the case of the UD57 sample, the fits of the $\Delta \sigma_H(H,T)$ curves can only be performed for temperatures above the mean field temperature $T_{c0}$ using the value of $T_{c0} = 72$K in the ALO expression. It can be
seen in Table 1 that the value of $H_{c2}$ extracted from the low-field part of the magnetoconductivity data matches very well that of $H'_c(0)$ obtained in a completely different way. Let us emphasize here that the comparison can only be indicative as the use of Eq.4 is not granted. So the value of $H'_c(0)$ could as well be a lower bound of the field above which the SCFs are suppressed. However, the fact that both fields are comparable for all the doping contents investigated highlights the consistency of our data analysis.

Table 1. Values of $H_{c2}(0)$ extracted from the fluctuation magnetoconductivity. They are comparable to the extrapolated values of the onset field of SCF $H'_c(0)$.(from ref.[7])

<table>
<thead>
<tr>
<th>Sample</th>
<th>UD57</th>
<th>UD85</th>
<th>OPT93.6</th>
<th>OD92.5</th>
</tr>
</thead>
<tbody>
<tr>
<td>$H_{c2}(0)(T)$</td>
<td>90(10)</td>
<td>125(5)</td>
<td>180(10)</td>
<td>200(10)</td>
</tr>
<tr>
<td>$H'_c(0)(T)$</td>
<td>86(10)</td>
<td>115(5)</td>
<td>155(10)</td>
<td>207(10)</td>
</tr>
</tbody>
</table>

The important result here is to show that the superconducting gap which is directly related to $H_{c2}(0)$ increases smoothly with increasing hole doping from the underdoped to the overdoped regime, contrary to the pseudogap which decreases. This is a strong indication that the gap determined here can thus be assimilated to the ”small” gap detected recently by different techniques, while the pseudogap would be rather connected with the ”large” gap [5].

One can point out that rather different values of $H_{c2}$ have been reported in ref.[21] from the analysis of the $T$ dependence of the magnetoconductivity at 1T in untwinned YBCO crystals. Even if this study was performed on different single crystals, one can conjecture that the very few data points used in that case to fit the fluctuation magnetoconductivity in a temperature range where the validity of the GL approach was not really checked, might be the source of large errors in the determination of $H_{c2}$.

6. Influence of disorder

It is now well admitted that the properties of cuprates are strongly dependent on disorder. We have studied for long the effect of the introduction of controlled disorder by electron irradiation and the way it affects the transport properties [22]. In particular, we have shown that similar upturns of the low-T resistivity are found for controlled disorder in YBCO and in some ”pure” low-$T_c$ cuprates, which indicates the existence of intrinsic disorder in those families [23].

We have also carried out magnetoresistance measurements in some OPT93.6 and UD57 samples irradiated by electrons. When $T_c$ is decreased by disorder, we find that both $T'_c$ and $H'_c(0)$ are also affected. The reduction in $T'_c$ nearly follows that in $T_c$ for the underdoped sample while it is slightly larger for the OPT sample. Consequently, when $T_c$ is decreased by disorder, the relative range of SCFs with respect to the value of $T_c$ expands considerably. For instance, in ref.[7], we still detect $T'_c \sim 60K$ in an UD57 irradiated sample with $T_c = 4.5K$.

These results allow us to draw important conclusions on the cuprate phase diagram. Indeed, contrary to $T_c$, $T'_c$ or $H'_c$, the pseudogap temperature $T^*$ has been found very early to be quite robust to disorder [24]. This is another indirect evidence that the pseudogap phase is not related to superconductivity. We want also to emphasize here that specific effects induced by disorder are probably at the origin of many confusions in the study of high-$T_c$ cuprates. This leads us to propose in fig.10 a 3D phase diagram where the effect of disorder has been introduced as a third axis.

There, in the pure systems, the occurrence of SCFs and the difficulty to separate the SC gap from the pseudogap in zero-field experiments justifies that the $T_c$ line could often be taken as a
Figure 10. Phase diagram constructed on the data points obtained here, showing the evolution of $T'_c$, the onset of SCF, with doping and disorder (from ref. [7]). The fact that the pseudogap and the SCF surfaces intersect each other near optimum doping in the clean limit is apparent. These surfaces have been limited to experimental ranges where they have been determined experimentally.

continuation of the $T^*$ line.

It can also be seen in this figure that the respective evolutions with disorder of the SC dome and of the amplitude of the SCF range explains the phase diagram often shown in a low-$T_c$ cuprate such as Bi-2201 and sketched in fig. 1(a). Finally, for intermediate disorder, the enhanced fluctuation regime with respect to $T_c$ observed in the Nernst measurements for the La$_{2-x}$Sr$_x$CuO$_4$ can be reproduced as well [4].

7. Conclusion

We have presented here a condensed report of our quantitative study of the superconducting fluctuation conductivity in YBCO that was extensively detailed in previous publications [6]-[8]. Our data give important determinations of some thermodynamic properties of the SC state of high-$T_c$ cuprates which are not accessible otherwise, as flux flow dominates near $T_c$ in the vortex liquid phase and the highest fields available so far are not sufficient to overcome $H_{c2}$ and then to reach the normal state at $T = 0$.

The consistency of our data analyses, which establish that the SCFs do match quantitatively the expectations from the 2D GL approach, strongly justifies the method used to determine the superconducting fluctuation conductivity from the deviations of the magnetoresistance from the $H^2$ normal state observed well above $T_c$. In a metallic state, deviations of the MR from an $H^2$ behavior could of course be expected in case of Fermi surface reconstruction. A two-carrier model has been indeed proposed to explain some MR data in YBa$_2$Cu$_{4}$O$_{8}$ [25] in which quantum oscillations could be detected at low $T$. In that approach the contributions of the SCFs has been completely ignored above $T_c$, though they should undoubtedly be present.

For our three higher oxygen content samples around optimal doping, Fermi Surface reconstructions have never been observed so far. As for our underdoped sample, it has a lower doping and lower $T_c$ than the ortho-II ordered YBCO$_{6.5}$ sample in which Fermi surface reconstruction has been detected at the highest $T$ of about 60K [26]. Our UD sample being twinned and thus with poor oxygen order, a reconstruction, if any, should then occur at even lower $T$. So, if such reconstruction effects do occur in this very sample, their influence should not extend in the high temperature range of our measurements (from 70K up to 130K). The oxygen disorder could play a role in reducing the $T_c$ of that sample. As in our lower $T_c$ samples in which disorder has been introduced on purpose, this could justify that, in presence of disorder, we need to introduce a mean field $T_c$ value higher than the zero field $T_c$ to interpret the data in
a GL approach \[7\].

Finally, contrary to what was claimed by others in this conference \[27\], our data unambiguously show that the superconducting fluctuations vanish abruptly with increasing temperature, allowing us to define an onset temperature $T'_c$ and an onset magnetic field $H'_c$ above which the SCF contribution to conductivity becomes unmeasurable. The comparison between the huge drop of the electronic susceptibility determined by NMR from $T^*$ and the emergence of superconducting fluctuations at $T'_c$ well below $T^*$ clearly indicates that these two temperature scales are not connected with each other.

We therefore have evidenced that the fluctuation conductivity can be very well accounted for in terms of the first series expansion of gaussian fluctuations in a limited temperature range, but require extension of the theory to explain the sharp drop at higher $T$. Moreover, the analysis of the fluctuating magnetoconductivity in this temperature range allows us to demonstrate that the pairing energy and SC gap both increase with doping, confirming then that the pseudogap has to be assigned to an independent magnetic order or crossover due to the magnetic correlations.

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[27] Taillefer L. Chang J et al. talks at this M2S conference