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DECISION of 20 June 2002

Case Number:

T 0977/98 - 3.4.3

Application Number:

88902345.3

Publication Number:

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IPC:

C01F 11/02

Language of the proceedings: EN

Title of invention:

Superconductivity in square-planar compound systems

Patentee:

UNIVERSITY OF HOUSTON

Opponent:

International Business Machines Corporation (IBM) Hoechst AG

Headword:

Ya-Ba-Cu-O superconductor/HOUSTON

Relevant legal provisions:

EPC Art. 87(1), 54, 56

RPBA Art. 9(2)

Keyword:

"Cases T 0976/98 and T 0977/98 consolidated"

"Priority: enabling disclosure in priority document"

"Product parameter (oxygen content) not directly derivable in priority document"

Decisions cited:

G 0002/98, G 0009/91

Catchword:



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Boards of Appeal

Chambres de recours

Case Number: T 0977/98 - 3.4.3

D E C I S I O N of the Technical Board of Appeal 3.4.3 of 20 June 2002

Appellant I:

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Decision under appeal:

Interlocutory decision of the Opposition Division of the European Patent Office posted 27 July 1998

concerning maintenance of European patent No. 0 341 266 in amended form.

Composition of the Board:

Chairman:

Members:

R. K. Shukla G. L. Eliasson M. J. Vogel

## Summary of Facts and Submissions

I. Appeal T 976/98 lies from the interlocutory decision of the opposition division dated 27 July 1998 proposing to maintain European Patent No. 0 280 812 in amended form pursuant to Article 102(3) EPC.

Appeal T 977/98 lies against the interlocutory decision of the opposition division dated 27 July 1998 proposing to maintain the European patent No. 0 341 266 in amended form following the opposition proceedings which were consolidated with that in respect of the abovementioned patent No. 0 280 812.

The European patents Nos. 0 280 812 and 0 341 266 have identical subject matter and were to be maintained in identical form. Pursuant to Article 9(2) RPBA, appeals T 976/98 and T 977/98 were consolidated with the consent of the parties; these being the same for both the appeal cases.

- II. The independent claims 1, 4, 8, and 11 as granted read as follows:
  - "1. A method of conducting an electrical current within a conductor material without substantial electrical resistive losses, characterized by the steps of:

utilizing as the conductor material a metal oxide complex of the general formula:

 $[L_{1-x}M_x]_aA_bO_v$ 

wherein "L" is scandium, yttrium, lanthanum, lutetium, or a combination thereof; "M" is barium, strontium, calcium, magnesium or a combination thereof provided that when "L" is lanthanum, "M" is not barium, strontium or calcium; "A" is copper; "x" is from 0.01 to 0.5; "a" is 1 to 2;

"b" is 1; and "y" is 2 to 4 that provides the metal oxide complex with substantially zero electrical resistance at a temperature of 77 K;

cooling said metal oxide complex to a temperature at or below that at which said metal oxide complex becomes superconductive; and

initiating a flow of electrical current within said metal oxide complex while maintaining said metal oxide complex at or below the temperature at which it becomes superconductive."

"4. A method of conducting an electrical current within a conductor material without substantial electrical resistive losses, characterized by the steps of:

utilizing as the conductor material a L-M-A-O complex wherein "L" is yttrium, lanthanum, scandium, lutetium, or a combination thereof; "M" is barium; "A" is copper,

said complex comprising a superconductive crystalline phase having a crystal structure uncharacteristic of that of a  $K_2NiF_4$  structure and having substantially zero electrical resistance at 77 K,

said superconductive crystalline phase having a formula  $L_1M_2A_3O_{6+\delta}$  wherein  $\delta$  has a number value of 0.1 to 1.0."

"8. A composition which is superconductive at a temperature of 77 K, characterized by:

a sintered metal oxide complex of the formula  $(L_{1-x}M_x)_aA_bO_y$  wherein "L" is scandium, yttrium, lanthanum, lutetium, or a combination thereof; "M" is barium, strontium, calcium, magnesium or a combination thereof provided that when "L" is lanthanum, "M" is not barium, strontium or calcium; "A" is copper; "x" is from 0.01 to 0.5; "a" is 1 to 2; "b" is 1; and "y" is 2 to 4."

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"11. A L-M-A-O complex wherein "L" is yttrium, lanthanum, scandium, lutetium, or a combination thereof; "M" is barium; "A" is copper,

said complex comprising a superconductive crystalline phase having a crystal structure uncharacteristic of that of a  $K_2NiF_4$  structure and having substantially zero electrical resistance at 77 K,

said superconductive crystalline phase having a formula  $L_1M_2A_3O_{6+\delta}$  wherein  $\delta$  has a number value of 0.1 to 1.0."

- III. The claims as proposed for maintenance in the decision under appeal differ from those as granted only in that the term "a L-M-A-O complex" in independent claims 4 and 11 is replaced by "a L-M-A-O complex comprising multiple phases".
- IV. In the opposition proceedings, the following documents, among others, were cited:
  - E3: Priority document US 12205/1987, filed on 6 February 1987, of the patent in suit;
  - E4: Priority document US 32041/1987, filed on 26 March 1987, of the patent in suit;
  - E6: Chemtech, 1987, vol. 17, pages 542 to 551;
  - E7: Physical Review Letters, vol. 58, No. 9, 1987, pages 908 to 910;
  - E8: EP-A-0 281 753;
  - E8a: Priority document US 24653/1987, filed on 11 March 1987, of document E8;

E9: Solid State Communications, vol. 66, 1988, pages 953 to 959;

E12: Physical Review B, vol. 36, No. 10, 1 October 1987, pages 5731 to 5734; and

E14: EP-A-0 274 407.

- V. In the decision under appeal, the opposition division reasoned essentially as follows:
  - (a) According to the description, the term "complex" in the claims under consideration has the meaning of a sample.
  - (b) Independent claims 4 and 11 of the patent as granted specify a complex comprising a superconductive crystalline phase having a formula  $L_1M_2A_3O_{6+\delta}$  (in the following referred to as the "123-phase"). Thus, claims 4 and 11 include within their scope a sample consisting only of this specific 123-phase.
  - (c) The priority document E3 does not, however, provide an enabling disclosure for a sample consisting of the 123-phase: At the filing date of the priority document E3, the cause for the superconductivity at 77 K was not clear. The skilled person therefore would have had to find out how many phases were present in the sample, and to determine the role of the different phases in superconductivity. Moreover, the priority document E3 teaches to use a rapid quenching step which in all probability would fail to produce a sample which is superconducting at 77 K. Therefore, the skilled person would additionally have to solve the problem of finding an optimized process for producing a sample which is

superconducting at 77 K starting with the composition disclosed in the priority document E3. Since at the filing date of the priority document E3, no well-established general level of knowledge in the field of high-temperature oxide superconductors existed, and since superconductivity of the 123-phase depends upon its oxygen content and the process of manufacture, the skilled person starting from the priority document E3 would have to invent a suitable process to produce a sample consisting of the 123-phase which is superconducting at 77 K.

Therefore, independent claims 4 and 11 of the patent as granted are not entitled to the priority date of 6 February 1987, but are entitled to the later priority date of 26 March 1987 derived from the priority document E4.

- (d) European patent application E8 claims a priority date of 11 March 1987 from priority document E8a which lies before the priority date of 26 March 1987 of the fourth priority document E4 of the patent in suit. Similarly, document E14 claims a priority date of 10 March 1987. Both the documents are thus comprised in the state of the art pursuant to Article 54(3) EPC. Each of these documents discloses samples comprising the 123-phase of Y-Ba-Cu-O and being superconducting at 77 K. Thus, independent claims 4 and 11 as granted are not novel.
- (e) The priority document E3 of the patent in suit dated 6 February 1987 provides an enabling disclosure for producing a sample (i.e. "complex") comprising multiple phases, since a skilled person starting from the teaching of document E3 would,

by carrying out routine experimentation, inevitably arrive at a sample comprising, among other phases, the superconducting 123-phase.

Therefore, claims 4 and 11 restricted to a L-M-A-O complex comprising multiple phases are entitled to the priority date of 6 February 1987.

Consequently, documents E8 and E14 do not belong to the state of the art under Article 54(3) EPC.

Also having regard to the other cited documents, the claims involve an inventive step.

- VI. The patent proprietor, University of Houston (appellant I), and Opponent II, Hoechst AG (appellant II), lodged appeals on 2 October 1998, both paying the appeal fees the same day. Both appellants filed statements of the grounds of appeal on 7 December 1998. Opponent I, IBM, is a party to the appeal proceedings as of rights (Article 107 EPC).
- VII. The following documents, inter alia, were cited by the opponents in the appeal proceedings:

E14a: Priority document US 1682/87 of 9 January 1987 for document E14;

E14b: Priority document US 21229/87 of 3 March 1987 for document E14;

E14c: Priority document US 24046/87 of 10 March 1987 for document E14; and

E27: R. M. Hazen, The Breakthrough -- The Race for the Superconductor (Summit Books, New York, 1988), pages 60, 72 to 74, 144 to 148.

VIII. At the oral proceedings held on 20 June 2002, the patent proprietor filed new claims forming first to third auxiliary requests. The parties made the following requests:

> The patent proprietor (appellant I) requested that the decision under appeal be set aside and that the patent be maintained according to anyone of the following requests:

Main request:

patent as granted;

First auxiliary request: on the basis of claims 1 to 14 according to the first

auxiliary request;

Second auxiliary request: on the basis of claims 1 to

14 according to the second

auxiliary request;

Third auxiliary request: on the basis of claims 1 to 6

according to the third auxiliary request.

The Opponent II (appellant II) requested that the decision under appeal be set aside and that European patent No. 0 341 266 be revoked.

IX. Claims 4 and 11 according to the first auxiliary request differ respectively from those as granted in that  $\delta$  is specified to have a number value of 1.0. Claims 1 to 3, 5 to 10 and 12 to 14 are the same as granted.

> Claims 4 and 11 according to the second auxiliary request differ respectively from those as granted in that  $\delta$  is specified to have a number value 1.0, and

that "a L-M-A-O complex" is replaced by "a L-M-A-O complex comprising multiple phases". Claims 1 to 3, 5 to 10 and 12 to 14 are the same as granted.

Claims 1 to 3 and 4 to 6 according to the third auxiliary request correspond to claims 1 to 3 and 8 to 10, respectively, of the claims as granted.

- X. The patent proprietor (appellant I) presented essentially the following arguments in support of his requests:
  - Example XI of the priority document E3 provides an (a) enabling disclosure for claims 4 and 11 according to the main request, and is in particular enabling disclosure for a sample consisting of the superconducting phase having the composition  $L_1M_2A_3O_{6+\delta}$  where  $\delta$  has a numerical value of 0.1 to 1.0 (the 123-phase). The opposition division has arrived at their decision in the mistaken belief that in order to manufacture a sample consisting of the 123-phase, it would have been necessary to use the method of document E8. There is however an important difference between manufacturing a bulk superconductor and isolating the superconducting phase from the multiple phases produced according to the method of Example XI. It was known in the art to separate the superconducting phase from non-superconducting phases by grinding the sample and using the Meissner effect.
  - (b) Contrary to the contentions of the opponents, the sample produced in Example XI of the priority document E3 contains a significant amount of the 123-phase thus facilitating the separation of the superconducting phase. From a simple stoichiometric calculation it is evident that 26 weight % of the sample in Example XI of the

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priority document E3 was of the 123-phase, which is in contrast to the submission in the decision under appeal alleging that only 2% was superconducting.

- (c) As known today, the black 123-phase consists of two phases, a superconducting, orthorhombic phase with the composition  $Y_1Ba_2Cu_3O_7$ , i.e.  $\delta = 1$ , and a non-superconducting, tetragonal phase with the composition  $Y_1Ba_2Cu_3O_6$  ( $\delta = 0$ ). A compound having the formula Y1Ba2Cu3O6+5 is thus a mixture of the orthorhombic and tetragonal phases in the proportions determined by the value of  $\delta$ . Starting from the teaching of Example XI of the priority document E3 and using known method for separating the superconducting phase from the nonsuperconducting phases, it was possible for the skilled person to produce superconducting samples having the composition  $Y_1Ba_2Cu_3O_{6+\delta}$  and having values of the oxygen parameter  $\delta$  in the whole range of 0.1 to 1.0.
- (d) Document E6 provides evidence that within a few days from obtaining a preprint of document E7, which was written by the inventor and discloses essentially the same subject matter as Example XI of the priority document E3, several laboratories around the world not only produced mixed phase Y-Ba-Cu-O superconducting samples, but also succeeded in isolating and identifying the superconducting 123-phase, although the disclosure in document E7 is less detailed than that in Example XI of the priority document E3.
- (e) It is taught in Example XI of the priority document E3 that after heating the compressed powder, the sample is "quenched". From the entire disclosure of document E3, it is evident that

"quenching" means "cooling in air", and not that the sample is immersed in a cold liquid, such as liquid nitrogen, as contended by Opponent II.

- (f) Although the patent in suit contain the incorrect information that the superconducting phase is tetragonal, it is submitted that document E14 is also misleading in the same respect (cf. E14c, page 15, lines 3 and 4).
- (g) Neither document E8 nor E14 provides an explicit disclosure of a combination of process parameters which would lead to a value of  $\delta$  falling within the claimed range of 0.1 to 1.0. Therefore, documents E8 and E14 are not relevant.
- XI. The arguments put forward by Opponent II (appellant II) can be summarized as follows:
  - At the filing date of priority document E3, it was (a) not known whether a single or several phases were responsible for the superconductivity. Therefore, the skilled person starting from the teaching of Example XI document E3 had no idea about the isolation of the superconducting phase, in particular since the method described in Example XI is not favourable for producing a superconducting sample. It prescribes a very short annealing time (about 15 minutes), and includes the step of subjecting the sample to a step of "quenching" or even "rapid quenching". On the basis of the general knowledge which was gained subsequently (see e.g. documents E9 and E12), short annealing time and rapid quenching are exactly the opposite to what is required in order to obtain a satisfactory superconducting sample.

Therefore, the skilled person starting from the method described in Example XI would most likely fail to end up with a sample exhibiting superconductivity at 77 K.

- (b) The priority document E3 does not contain any disclosure from which one can directly and unambiguously derive the range of the oxygen parameter δ from 0.1 to 1.0 which is specified in independent claims 4 and 11 according to the main request. The only disclosure is a single sample in Example XI. It is not evident how one sample can provide support for a whole range of the oxygen parameter δ. Following the criteria set out in G 2/98, the subject matters of independent claims 4 and 11 according to the main request and the first and second auxiliary requests are thus not entitled to the priority date based on document E3.
- (c) It follows from Figure 2 of document E9 that for compositions with the oxygen parameter  $\delta$  less than about 0.7, the 123-phase having the composition  $Y_1Ba_2Cu_3O_{6+\delta}$  is no longer superconducting at 77 K, and for  $\delta$  less than about 0.4, it is not superconducting at all. Thus, for most part of the claimed range of  $\delta$  from 0.1 to 1.0, the composition  $Y_1Ba_2Cu_3O_{6+\delta}$  fails to meet the requirement of being superconductive at 77 K. Therefore, apart from not having any basis in the priority document E3, the lower limit of 0.1 is meaningless.
  - (d) It is furthermore shown in document E9, as well as in document E12, that the composition  $Y_1Ba_2Cu_3O_{6+\delta}$  goes through an tetragonal/orthorhombic phase transition at  $\delta$  having a value of about 0.35. Thus, the model suggested by the patent proprietor

that  $Y_1Ba_2Cu_3O_{6+\delta}$  is a mixture of tetragonal  $Y_1Ba_2Cu_3O_6$  and orthorhombic  $Y_1Ba_2Cu_3O_7$  has no basis. Furthermore, since the critical temperature  $T_c$  is nearly constant in the range of  $\delta$  between 0.9 and 1.0, it would be difficult, if not impossible, for the skilled person to separate the phase corresponding to  $\delta=1$ . The patent in suit also provides misleading information by stating that the superconducting 123-phase is tetragonal instead of orthorhombic, and that the preferred range of  $\delta$  is between 0.1 and 0.5, i.e. completely outside of the range where the 123-phase is superconducting (cf. patent, page 7, lines 14 to 15).

- (e) Document E14 discloses all features of independent claims 4 and 11 according to the main request, since the composition YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6.9</sub> is explicitly disclosed therein. Moreover, the priority document E14c contains an enabling disclosure for producing a sample having this composition (cf. page 8, lines 10 to 13; page 13; page 4, line 30 in combination with page 8).
- XII. The arguments of the non-appealing Opponent I can be summarized as follows:
  - (a) The great difficulties the patent proprietor had in progressing beyond the multi-phase sample of Example XI in E3 is documented in E27 written by Robert Hazen, a member of the inventor's research team. Thus, it was not as straight-forward as the patent proprietor alleges to identify and separate the 123-phase.
  - (b) The black phase of the Y-Ba-Cu-O sample produced according to the method of Example XI was subsequently identified as perovskite-related

tetragonal phase. However, it is the orthorhombic phase of the black material which is superconducting.

- (c) The rapid quench step described in Example XI of document E3 would produce only the tetragonal 123-phase which is not superconducting, since only the orthorhombic 123-phase is superconducting.
  - (d) Although the team of IBM managed to reproduce the results presented in document E7 in a very short time (four days), the number of man-hours was enormous. Furthermore, proprietary in-house knowledge of IBM and not routine knowledge was crucial for making it possible to produce and identify the superconducting 123-phase starting from the composition disclosed in document E7.

#### Reasons for the Decision

1. Both appeals meet the requirements of Articles 106 to 109 and Rule 64 EPC and are therefore admissible.

#### 2. Amendments

Amendments in relation to claims as granted have been made to independent claims 4 and 11 of the first and second auxiliary requests. There were no objections pursuant to Article 123(2) and (3) EPC against the amendments from the opponents, and the Board is also satisfied that the amendments comply with the requirements of Article 123(2) and (3) EPC.

- 3. Priority main request and first and second auxiliary requests
- 3.1 The patent in suit claims four priority dates, i.e.
  12 January 1987, 27 January 1987, 6 February 1987, and
  26 March 1987. It is the priority date of 6 February
  1987 claimed from the priority document E3 for
  independent claims 4 and 11 of the main request and
  first and second auxiliary requests, which is in
  dispute in the present appeal. In this connection, the
  opponents contended that the document E3 did not
  contain an enabling disclosure to realize the invention
  as claimed in claims 4 and 11, so that the invention as
  claimed was not the same as the one disclosed in the
  priority document E3, contrary to the requirements of
  Article 87(1) EPC.

Referring to the decision G 2/98, the opponents furthermore argued that claims 4 and 11 were not entitled to the priority date of document E3, since the priority document discloses neither the formula  $L_1 M_2 A_3 O_{6+\delta}$  nor the numerical values of the oxygen parameter  $\delta$  as specified in claims 4 and 11.

The disclosure in the priority document E3 which is relevant to claims 4 and 11 is the Example XI. It is reproduced as Example XI in the patent in suit (cf. page 12, lines 20 to 47). Example XI describes a method of producing a Y-Ba-Cu-O sample, termed "complex" in the patent in suit (cf. item V(a) above), starting from a mixture of Y<sub>2</sub>O<sub>3</sub>, BaCO<sub>3</sub>, and CuO having the nominal composition Y<sub>1.2</sub>B<sub>0.8</sub>CuO<sub>y</sub>. After mixing the components, the oxide mixture was compressed to pellets at an applied pressure of 100 to 500 psi, followed by heating the pellets in air at 900 to 1100°C for about 15 minutes and then "rapidly quenching" the pellets to room temperature in air.

The Y-Ba-Cu-O sample produced according to the above method was shown to comprise multiple phases where at least one of the phases has a structure uncharacteristic of that of a K<sub>2</sub>NiF<sub>4</sub> crystal and exhibited a superconducting transition between 80 and 93 K. At a temperature of 4.2 K, the sample showed a diamagnetic signal corresponding to 24% of the superconducting signal of a lead sample with similar dimensions.

3.3 According to established case law of the boards of appeal, if a method of producing a substance is disclosed, then the product resulting from the disclosed method is also disclosed, provided that the method is an enabling disclosure, i.e. a disclosure sufficiently clear and complete for it to be carried out by a skilled person. When the resulting product is in form of multiple phases, each of the phases resulting from the disclosed method are considered to be disclosed, if separation and identification of the individual phases can be carried out in a routine manner using the techniques available at the date in question.

In the present case, the Y-Ba-Cu-O sample produced by the method disclosed in Example XI contains multiple phases. The superconducting phase was not isolated but was only identified to have a crystal structure uncharacteristic of that of a  $\rm K_2NiF_4$  crystal and to have a superconducting transition between 80 and 93 K.

3.4 The arguments presented by the opponents as to why Example XI of the priority document E3 could not be considered to be an enabling disclosure were essentially as follows (cf. items XI(a) and XII(a) to (d) above): Firstly, the method of Example XI of the priority document E3 suggests a set of process parameters, in particular the short heating step and

the rapid cooling (quenching) of the sample, which are so unfavorable that the skilled person following the teaching of Example XI would inevitably fail to produce an Y-Ba-Cu-O oxide superconductor having a superconducting transition above 77 K.

Secondly, claims 4 and 11 cover the case where the L-M-A-O sample consists of a superconductive phase having substantially zero electrical resistance at 77 K. The priority document E3, according to the opponents, does not provide an enabling disclosure for this case, since the method according to Example XI would at most produce a mixed phase Y-Ba-Cu-O sample, and not a single superconducting phase sample. It was not routine to separate and identify the superconducting phase in order to arrive at a sample consisting of the superconducting phase. Since the cause of the superconductivity was not known, the skilled person had no idea whether one or several phases were responsible for the superconductivity.

3.4.1 As regards the relatively short heating time of about 15 minutes and the cooling rate ("rapid quenching") employed in Example XI of the priority document E3, as accepted by the patent proprietor, these are not optimum process parameters. However, the Board accepts the patent proprietor's submission that the quenching employed in documents E6 (cf. Figures 6 to 8) and E12 (cf. page 5732, "Experimental Procedure") entailed quenching by immersing the sample directly in liquid nitrogen (cf. E9, page 954, left column, second paragraph), whereas rapid quenching employed in the priority document E3 was carried out by cooling in air. Consequently, in the Board's view, it was doubtful whether the results reported in documents E6 and E12 were applicable to the method reported in the priority

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document E3, and whether they conclusively show that a sample produced by quenching by cooling in air as in the priority document E3 would not contain the superconducting 123-phase.

- Regarding the second argument that it was not routine 3.4.2 to separate the superconducting phase from the nonsuperconducting phases, the Board is convinced by the patent proprietor's arguments that at the priority date of document E3, it was possible to separate and identify the superconducting phase, later known as the "123-phase", from the non-superconducting phases using standard techniques which were available at that time (cf. item X(a) above). The superconducting phase could be isolated by, for example, grinding the sample and employing the Meissner effect whereby the superconducting grains are separated from the nonsuperconducting grains in a magnetic field. Furthermore, the exact crystal structure of the superconducting phase could be determined using x-ray diffraction. Although these techniques may be tedious and time-consuming, they were nevertheless regarded as routine at the priority date of document E3.
- that within a couple of weeks from learning about the present invention through the publication of document E7, numerous laboratories all over the world not only managed to reproduce the results of document E7, but also succeeded in isolating, identifying, and characterizing the superconducting 123-phase, although document E7 describes the fabrication process in less detail than in Example XI of the priority document E3 (cf. E6, page 545, second paragraph; E27, page 72, last paragraph to page 74, as well as the argument of the patent proprietor, items X(a) and (b) above).

Thus, taking the above facts and arguments into consideration, the Board concludes that the method of Example XI was disclosed in sufficient detail in order to enable a skilled person to produce a Y-Ba-Cu-O sample comprising a superconductive crystalline phase having a crystal structure uncharacteristic of that of a  $K_2NiF_4$  crystal and having substantially zero electrical resistance at 77 K.

- Claims 4 and 11 specify in addition that the superconductive crystalline phase has a formula  $L_1M_2A_3O_{6+\delta}$ . For the main request, the oxygen parameter  $\delta$  is set to have a numerical value of 0.1 to 1.0, whereas for the first and second auxiliary requests, the value of the oxygen parameter  $\delta$  is fixed to 1.0. It was not disputed by the patent proprietor that the priority document E3 does not explicitly disclose the formula or any numerical value of the oxygen content of the superconducting phase.
- 3.5.1 Regarding the main request, Opponent II argued that the feature "having a formula  $L_1M_2A_3O_{6+\delta}$  where  $\delta$  has a number value of 0.1 to 1.0" is not directly derivable from the priority document E3, as required following the Enlarged Board decision G 2/98, since the disclosure of a single sample cannot provide support for a whole range of the oxygen parameter  $\delta$  (cf. item XI(b) above). Furthermore, for YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6+ $\delta$ </sub>, which is the superconducting phase relevant to the method of Example XI of the priority document E3, the lower limit of  $\delta$  = 0.1 is not superconducting at all, and YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6+ $\delta$ </sub> fails to be superconducting at 77 K for most part of the claimed range from 0.1 to 1.0 (cf. item XI(c) above).

As to the first and second auxiliary requests,  $\text{Opponent II argued that the claimed value of 1.0 for } \\ \text{the oxygen parameter } \delta \text{ is not directly derivable from }$ 

the priority document E3, since carrying out the process of Example XI would not inevitably result in a superconducting phase having the oxygen parameter  $\delta$  equal to 1.0.

- 3.5.2 The patent proprietor responded that the 123-phase of Y-Ba-Cu-O having the general formula  $Y_1Ba_2Cu_3O_{6+\delta}$  is actually a mixture of a superconducting orthorhombic 123-phase corresponding to  $\delta=1$ , and a nonsuperconducting tetragonal 123-phase corresponding to  $\delta=0$  (cf. item X(c) above). Therefore, it would have been possible for a skilled person, using known methods at the priority date E3, to separate the two 123-phases so as to produce samples having values of  $\delta$  falling within the whole range as claimed.
- 3.5.3 Although the Board is convinced that a skilled person following the teaching of Example XI in the priority document E3 would be able to establish the composition YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6+ $\delta$ </sub> for the superconducting phase using routine techniques, the Board agrees with Opponent II that the priority document E3 does not contain any implicit disclosure of the end values 0.1 and 1.0 for the oxygen parameter  $\delta$ , since the skilled person is not taught a specific combination of process conditions which would inevitably lead to these particular values of the oxygen parameter  $\delta$ . Consequently, the priority document E3 cannot be regarded as unambiguously disclosing the same superconducting composition with  $\delta$  = 0.1 and  $\delta$  = 1.0 as the one claimed in claims 4 and 11 of the main request and first and second auxiliary requests.
- 3.5.4 The patent proprietor's argument that the superconducting 123-phase is a mixture of two phases having compositions with  $\delta=0$  and  $\delta=1.0$ , respectively, fails to convince the Board, since, as Opponent II pointed out, it is in contradiction with the findings disclosed in documents E9 and E12 (cf.

XI(d) above). Documents E9 and E12 are both published after all the priority dates of the patent in suit, and are therefore not comprised in the state of the art. However, they provide information about the material properties of Y,Ba,Cu,O<sub>6+5</sub>. The documents E9 and E12 report that when the oxygen parameter  $\delta$  falls below a value of about 0.4 to 0.5, an orthorhombic to tetragonal phase transition takes place and the superconductivity disappears (cf. E9, Figure 2; paragraph bridging pages 956 and 957; E12, paragraph bridging pages 5732 and 5733; as well as item XI(c) above). Above this value,  $Y_1Ba_2Cu_3O_{6+\delta}$  appears to be orthorhombic where the superconducting transition temperature  $T_c$  increases as  $\delta$  approaches the value of 1. These findings were not disputed by the patent proprietor.

Thus, the production and isolation of the superconducting phase having a composition with  $\delta$  equal to 1.0 is not as straightforward as the patent proprietor suggested under item X(c) above. Therefore, the Board is not convinced that the priority document E3 implicitly discloses the limits 0.1 and 1.0 for the oxygen parameter  $\delta$ , in the sense that a skilled person starting from document E3 would inevitably produce samples with varying values of  $\delta$  falling within the whole interval [0.1, 1.0] as claimed, and in particular with the endpoints 0.1 and 1.0.

Moreover, as Opponent II pointed out (cf. item XI(c) above), the lower limit of  $\delta$  = 0.1 corresponds to the tetragonal phase which is not superconducting at all. Therefore, it is highly unlikely that the superconducting phase produced according to the process in the priority document E3 had this specific composition.

3.5.5 According to G 2/98 (OJ EPO 2001, 413), priority of a previous application in respect of a claim in a European patent application is to be acknowledged only if the skilled person can derive the subject matter of the claim directly and unambiguously, using common general knowledge, from the previous application as a whole.

Since the priority document E3 does not implicitly disclose the end points 0.1, 1.0 for the oxygen parameter  $\delta$ , claims 4 and 11 according to the main request and the first and second auxiliary requests are not entitled to the priority date of E3.

4. Priority: third auxiliary request

The claims corresponding to the third auxiliary request correspond to claims 1 to 3 and 8 to 10 according to the main request. The opponents never challenged the finding in the decision under appeal that these claims are entitled to the priority date of E3, i.e.

6 February 1987. The Board is convinced that these claims are entitled to the priority date of E3.

5. Novelty and inventive step - main request, first and second auxiliary requests

Documents E7, E8, and E14 were considered most relevant by the parties in the appeal proceedings.

Document E7 is an article published on 2 March 1987 before the priority date of 26 March 1987 of the patent in suit. This document is therefore comprised in the state of the art pursuant to Article 54(2) EPC for independent claims 4 and 11 according to the main request and first and second auxiliary requests. It discloses the formation of a superconducting, mixed phase Y-Ba-Cu-O sample using powders of Y,O3, BaCO3, and

CuO in the nominal composition of  $Y_{1.2}Ba_{0.8}CuO_{4-\delta}$  as starting materials, which are mixed, compressed, and sintered (cf. paragraph bridging pages 908 and 909). The resulting sample comprises phases having a crystal structure uncharacteristic of that of  $K_2NiF_4$  and has a superconducting transition between 80 and 93 K (cf. page 908, last paragraph to page 909, penultimate paragraph). Thus, document E7 describes the same process but in less detail than in Example XI of the priority document E3 of the patent in suit (cf. item 3.2 above).

5.2 Document E8 is a European patent application claiming a priority date of 11 March 1987. Thus, for independent claims 4 and 11 according to the main request and the first and second auxiliary requests, document E8 is an earlier European application within the meaning of Article 54(3) EPC.

Document E8 discloses various samples having compositions very close to YBa<sub>2</sub>Cu<sub>3</sub>O<sub>y</sub> which are superconducting at 77 K (cf. column 1, line 53 to column 2, line 4; column 2, line 49 to column 3, line 17). It is furthermore taught in document E8 that the samples should be cooled slowly in oxygen in order to allow them to retain more oxygen (cf. column 2, lines 36 to 48). The corresponding disclosure is also found in the priority document E8a of 11 March 1987 (cf. page 3, line 26 to page 4, line 26).

Document E14 is a European patent application published on 13 July 1987 after the priority date of the patent in suit, and claims a priority of 10 March 1987 from the priority document E14c. Thus, for independent claims 4 and 11 according to the main request, and the first and second auxiliary requests, document E14 is an earlier European application within the meaning of Article 54(3) EPC.

Document E14 discloses the formation a Y-Ba-Cu-O sample having a superconducting phase with orthorhombic crystal symmetry which has the composition  $YBa_2Cu_3O_{6.9}$  (cf. page 4, lines 56 to 58; page 6, lines 30 to 59). The sample has zero resistance at a temperature  $T_c$  of 91.5 K (cf. Table, Example 1). The same information is found in the priority document E14c of 10 March 1987 (cf. page 8, lines 11 to 15; page 12, line 10 to page 13, line 27; Table, Example I).

### 5.4 Main request

5.4.1 As regards document E14, it was contended by the patent proprietor that applying the same standard of enabling disclosure as that for the priority document E3 (for the priority date of 6 February 1987) to the priority document E14c, document E14 was not entitled to the priority date of 10 March 1987, and therefore was not comprised in the state of the art pursuant to Article 54(3) EPC. In particular, it was argued that the process described in the priority document E14c, page 12, line 10 to page 13, line 27, does not specify a set of process parameters to produce a sample having a composition with the oxygen parameter δ equal to 0.9, so that there was no enabling disclosure in the priority document E14c.

In this connection, the Board observes that there is one crucial difference between the disclosures of both the priority documents. In document E3, neither the composition with the end values  $\delta=0.1$  and  $\delta=1.0$  of the oxygen parameter, nor process conditions which inevitably produce these compositions are disclosed. In document E14c on the other hand, the composition with  $\delta=0.9$  is specifically disclosed, so that the skilled person knew a priori the exact composition to be produced. The optimization to this end of starting

materials and the essential process conditions which are disclosed in document E14c, in the Board's view, therefore belong to the routine practice in the art.

The Board therefore concludes that the priority document E14c contains an enabling disclosure of the superconducting phase having a composition with the oxygen parameter  $\delta$  equal to 0.9. Document E14 is therefore entitled to the priority date of 10 March 1987 and thus belongs to the state of the art according to Article 54(3) EPC.

5.4.2 As mentioned above, document E14 discloses a superconducting phase a composition with the oxygen parameter  $\delta$  equal to 0.9. This value of the oxygen parameter  $\delta$  falls within the range of 0.1 to 1.0 as defined in claims 4 and 11. Therefore, the subject matters of independent claims 4 and 11 according to the main request are not new.

The main request is therefore not allowable.

- 5.5 First auxiliary request
- 5.5.1 The subject matters of independent claims 4 and 11 according to the first auxiliary request are new, since none of the cited documents E7, E8, and E14 disclose a superconducting phase having a composition with the oxygen parameter  $\delta$  equal to 1.0. Moreover, it is generally known in the art that the oxygen parameter  $\delta$  in the superconducting phase  $Y_1Ba_2Cu_3O_{6+\delta}$  is very sensitive to the choice of process parameters of the method of forming the superconductor, such as cooling rate and ambient oxygen content. Therefore, a skilled person carrying out the teaching of either document E7, E8 or E14 would not inevitably arrive at a sample having a superconducting phase with the oxygen parameter  $\delta$  equal to 1.

- 5.5.2 Since documents E8 and E14 are prior art falling under Article 54(3) EPC, they cannot be considered for inventive step. Thus, document E7 is the only remaining relevant prior art document.
- 5.5.3 The claimed subject matter has the advantage that the superconducting transition temperature is particularly high, and that other properties, such as critical current density, are improved over other phases having lower values of  $\delta$ . Thus, the objective problem relates to optimizing the superconductive properties of the Y-Ba-Cu-O sample, such as the transition temperature.
- 5.5.4 A skilled person would in the Board's opinion automatically seek to improve the properties of the mixed phase sample disclosed in document E7. As discussed under items 3.4.2 and 3.4.3 above, it appears from the parties' submissions that it was standard procedure to separate the superconducting phase from the other phases. Moreover, as evidenced from the numerous results from independent research laboratories within very short time from the publication date of document E7, it was evidently routine to change systematically the process parameters so as to maximize the transition temperature. By carrying out such routine experimentation, the skilled person would, in the Board's opinion, arrive at a sample comprising a superconducting phase having a composition with the oxygen parameter  $\delta$  equal to 1.
- 5.5.5 Therefore, in the Board's judgement, the subject matter of independent claim 11 according to the first auxiliary request does not involve an inventive step within the meaning of Article 56 EPC. The claims according to the first auxiliary request are therefore not allowable.

## 5.6 Second auxiliary request

Claim 11 according to the second auxiliary request differs from that of the first auxiliary request in that the L-M-A-O complex comprises multiple phases. Since this feature is known from document E7, the subject matter of claim 11 according to the second auxiliary request does not involve an inventive step for the same reasons as for the first auxiliary request.

#### 5.7 Third auxiliary request

An objection of lack of novelty against the claims 1 to 3 and 8 to 10 as granted, which now form the basis of the third auxiliary request, was raised by the Opponent II in the statement of opposition having regard to document E14 on the assumption that the document belonged to the state of the art pursuant to Article 54(3) EPC. In the decision under appeal, the opposition division concluded that these claims were novel (cf. Reasons, item 5.2.4). The Opponent II filed the complete priority documents E14a to E14c with the statement of the grounds of appeal, but did not dispute the novelty of claims 1 to 3 and 8 to 10 and did not provide any arguments to substantiate such an objection. Therefore, the patentability of the claims according to the third auxiliary request was not in dispute in the appeal proceedings (cf. G 9/91, OJ EPO 1993, 408).

The description of the patent in suit needs to be adapted to the claims according to the third auxiliary request, so that the Board decides to remit the case to the opposition division for further prosecution.

# Order

# For these reasons it is decided that:

- 1. The decision under appeal is set aside.
- The case is remitted to the first instance for further prosecution on the basis of claims 1 to 6 according to the third auxiliary request.

The Registrar:

The Chairman:

R. Schumacher

R. K. Shukla

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