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D E C I S I O N
of 3 May 2000

Case Number: T 0173/98 - 3.3.5

Application Number: 89903216.3

Publication Number: 0431160

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Language of the proceedings: EN

Title of invention:
Process for producing thin-film oxide superconductor

Patentee:
KABUSHIKI KAISHA TOSHIBA

Opponent:
Siemens AG

Headword:
-

Relevant legal provisions:
EPC Art. 56, 123(2)

Keyword:
"Inventive step (no) - obvious modification"
"Amendments - added subject-matter (yes)"

Decisions cited:
-

Catchword:
-

Case Number: T 0173/98 - 3.3.5

D E C I S I O N
of the Technical Board of Appeal 3.3.5
of 3 May 2000

Appellant: KABUSHIKI KAISHA TOSHIBA
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Representative: -

Decision under appeal: **Decision of the Opposition Division of the European Patent Office posted 19 November 1997 revoking European patent No. 0 431 160 pursuant to Article 102(1) EPC.**

Composition of the Board:

Chairman: G. J. Wassenaar
Members: A.-T. Liu
J. H. van Moer

Summary of Facts and Submissions

I. The appeal was from the decision of the opposition division revoking European patent No. 0 431 160 upon opposition filed against the patent on the grounds of Article 100(a), 100(b) and 100(c) EPC.

II. The European patent claimed a priority date of 16 March 1988 from Japanese application document JP 60308/88.

III. The decision was based on claims 1 to 21 of the main request and the first auxiliary request as filed with the letter dated 12 May 1997 and during the oral proceedings of 28 August 1997, respectively.

Claim 1 of this main request had been amended relative to claim 1 as granted. It read as follows:

"A method for manufacturing an oxide superconductor thin film, comprising the steps of:

preparing a substrate;

depositing a first thin film on said substrate; using a material containing constituent elements of said oxide superconductor thin film and supplied from a first source; and

supplying atomic oxygen, ozone or excited oxygen molecules to or near a thin film deposition site on said substrate during the deposition of said first thin film, so that oxygen atoms are taken into said first thin film to form said oxide superconductor thin film from said first thin film, said atomic oxygen, ozone or excited oxygen molecules being produced by causing an electric discharge in an oxygen gas or oxygen-containing mixed gas in a pipe or at the outlet of said pipe through which said gas is supplied from a second

source different from said first source, and the quantity of atomic oxygen, ozone or excited oxygen molecules supplied being sufficient to form said oxide superconductor thin film."

Claim 1 of the first auxiliary request differed from claim 1 of the main request essentially in the stipulation of the manner in which oxygen was incorporated and the gas discharged. The last part of this claim read as follows:

"... supplying atomic oxygen, ozone or excited oxygen molecules to or near a thin film deposition site on said substrate during the deposition of said first thin film, so that oxygen atoms are incorporated into the crystal structure of said first thin film to form said oxide superconductor thin film from said first thin film, said atomic oxygen, ozone or excited oxygen molecules being produced by causing an electric discharge in an oxygen gas or oxygen-containing mixed gas in a pipe or at the outlet of said pipe through which said gas is supplied from a second source different from said first source, the atomic oxygen, ozone or excited oxygen molecules being caused to flow as a jet from the pipe outlet to the region of the first thin film, and the quantity of atomic oxygen, ozone or excited oxygen molecules supplied being sufficient to form said oxide superconductor thin film; the method not including the introduction either (a) of oxygen ions as such or (b) an oxygen plasma to the first thin film."

IV. The opposition division accepted that a skilled person had sufficient knowledge to produce atomic oxygen, ozone and excited oxygen. The opposition division held

that there was, however, no teaching in the patent in suit on how to measure, calculate or otherwise deduce what is a "sufficient quantity" of these oxygen species as stipulated in claim 1 of both requests. The opposition division therefore concluded that the patent in suit did not fulfill the requirements of Article 100(b) EPC.

V. Reference was made to the following documents in the decision under appeal:

D10: EP-A-0 285 132

D12: Jap. J. Applied Physics 33, part 1, No. 78 (July 1994) pp. 4308-4311.

D13: J. Phys. D: Applied Physics 20 (1987), pp. 1421-1437.

VI. In appeal, the appellant submitted six sets of claims with the letter dated 1 April 2000. The claims of the main and first auxiliary request were the same as those on which the impugned decision was based. The remaining sets of claims were to form the basis for auxiliary requests 2 to 5.

Claim 1 of the second auxiliary request corresponded to claim 1 of the main request, with the difference that it no longer stipulated the gas discharge at the outlet of the pipe. The last part of claim 1 of this request read:

"... said atomic oxygen, ozone or excited oxygen molecules being produced by causing an electric discharge in an oxygen gas or oxygen-containing mixed

gas in a pipe through which said gas is supplied from a second source different from said first source, and the quantity of atomic oxygen, ozone or excited oxygen molecules supplied being sufficient to form said oxide superconductor thin film."

Likewise, claim 1 of the third auxiliary request corresponded to claim 1 of the first auxiliary request, with the difference that the gas discharge at the outlet of the pipe had been deleted from the claim. The last part of claim 1 thus read:

"... said atomic oxygen, ozone or excited oxygen molecules being produced by causing an electric discharge in an oxygen gas or oxygen-containing mixed gas in a pipe through which said gas is supplied from a second source different from said first source, the atomic oxygen, ozone or excited oxygen molecules being caused to flow as a jet from the pipe outlet to the region of the first thin film, and the quantity of atomic oxygen, ozone or excited oxygen molecules supplied being sufficient to form said oxide superconductor thin film; the method not including the introduction either (a) of oxygen ions as such or (b) an oxygen plasma to the first thin film."

Claim 1 of the fourth auxiliary request corresponded to claim 1 of the main request, with the difference that it further specified the method of discharge. To the wording of claim 1 of the main request was added:

"wherein the discharge is an RF discharge, high frequency discharge or a silent discharge".

Claim 1 of the fifth auxiliary request corresponded to

claim 1 of the second auxiliary request, with the same additional limitation regarding the method of discharge as for the fourth auxiliary request.

VII. During the oral proceedings which took place on 3 May 2000, the appellant submitted a set consisting of 25 claims as the basis for a new main request. The six previous requests filed on 1 April 2000 were correspondingly redefined as auxiliary requests 1 to 6.

Claim 1 of this final main request read:

"A method for manufacturing an oxide superconductor thin film, comprising the steps of:

preparing a substrate;

depositing an oxide superconductor thin film on said substrate; and

supplying atomic oxygen, ozone or excited oxygen molecules to or near a thin film deposition site on said substrate during the deposition of said thin film, said atomic oxygen, ozone or excited oxygen molecules being produced by means of generating a discharge in an oxygen gas or oxygen-containing mixed gas in a pipe through which the gas is supplied."

VIII. The following documents were introduced by the parties for the first time into the appeal proceedings:

D15: MBE Method using oxygen radical source (with an English translation), August 1992.

D16: Appl. Phys. Lett. 53 (18), p. 1762.

D17: J. Electronic Materials, vol. 16, No. 5, 1987, pp. 373 to 378.

IX. The appellant's arguments with respect to inventive step of the subject-matter according to claim 1 of the main request could be summarised as follows:

- The claimed process was essentially distinguished from the process of D16 in that the gas discharge took place in the oxygen supply pipe.

- The claimed process had the advantage of confining the highly reactive discharge gas, thereby reducing its contamination.

- D17 was directed to a process involving:

  (i) the oxidation of a formed wafer, and

  (ii) the production of SiO_2 films by this oxidation.

- In contrast to the process of claim 1, D17 thus did not concern:

  (i) the oxidation of a thin film during its formation, and

  (ii) the production of a superconductor due to the oxidation of the thin film.

- The skilled person looking for a solution to the problem of gas contamination in the technical field of superconductor thin films would not have considered applying the teaching of D17.

Concerning the wording "the quantity of atomic oxygen, ozone or excited oxygen molecules supplied being

sufficient to form said oxide superconductor thin film" in claim 1 of the auxiliary requests, the appellant submitted that this added feature could be derived from the application as originally filed for the following reasons:

- The claim did not preclude the presence of oxygen ions in the discharge gas.

- The concentration of oxygen ions was, however, much lower than that of neutral oxygen species.

- Proof that the ion concentration in the discharge gas was insignificant could be found in documents D12, D13 and D15.

- This view was in agreement with the finding in D16 that oxygen ions were detrimental to the process of forming superconductor thin films.

- The skilled person therefore knew that the formation of the superconductor thin film was due to neutral oxygen species being supplied in sufficient quantities for the required oxidation.

X. The respondent put forward the argument that D17 essentially disclosed a process and apparatus for oxidising a substrate with oxygen-containing discharge gas. Furthermore, it explicitly addressed the problem of contamination in connection with the method of gas discharge. This teaching was therefore valid, irrespective of the object to be oxidised with the discharge gas.

The respondent refuted the appellant's argument that

the ion concentration in the discharge gas was in principle insignificant and that the oxidation of the thin film was necessarily attributed to the neutral oxygen species in the discharge gas.

XI. At the end of the oral proceedings, the appellant (patentee) requested that the decision under appeal be set aside and that the patent be maintained on the basis of the main request submitted during the oral proceedings or, in the alternative, on the basis of any of the requests filed with letter of 1 April 2000 redefined as auxiliary requests 1 to 6.

The respondent (opponent) requested that the appeal be dismissed.

Reasons for the Decision

Main request

1. *State of the art*

Claim 1 of the main request is directed to a method comprising

(i) depositing a oxide superconductor thin film on a substrate and

(ii) supplying atomic oxygen, ozone or excited oxygen molecules to or near a thin film deposition site on said substrate.

The Board notes that claim 1 does not stipulate a particular method for depositing the thin film. The

claimed method thus encompasses the following elements:

(i) depositing a thin film using a method other than laser sputtering, and

(ii) supplying excited molecular oxygen during deposition.

These features are not disclosed in the priority document. The Board therefore finds that the priority document and present claim 1 do not concern the same invention. Consequently, claim 1 cannot enjoy the priority date. The effective filing date for the subject-matter of claim 1 is thus the filing date of the European patent application of the patent in suit, i.e. 15 March 1989. Consequently, document D16 bearing the publication date of 31 October 1988 forms part of the state of the art for the subject-matter of claim 1. This finding has not been disputed.

2. *Inventive step*

2.1 The Board considers that the closest prior art is represented by D16. This document discloses a plasma-assisted laser deposition (PLD) process for making $YBa_2Cu_3O_{7-x}$ superconductor thin films by focusing the laser onto the target in the presence of an O_2 discharge. The discharge is caused by a middle ring electrode positioned between the target and the substrate. The presence of atomic oxygen in the discharge gas is clearly observed by its emission line(s) (see page 1762, left-hand column, paragraph 2 to right-hand column, paragraph 3).

2.2 According to the patent in suit, the problem to be

solved by the invention is to provide a thin film which is superconducting as-deposited, without its being subjected to heat treatment after its formation (column 1, lines 36 to 53; column 3, line 50 to column 4, line 1). This problem is, however, already solved by the process of D16 (see page 1762, left-hand column, paragraph 1: "$YBa_2Cu_3O_{7-x}$ thin film deposited at 400°C ... did not require any post-annealing to be superconducting"; and page 1762, right-hand column, paragraph 4: "The films obtained were all superconducting as-deposited without post-annealing").

During the oral proceedings, the appellant advanced the argument that, when the gas is discharged in the manner as disclosed for the process of D16, the reactive oxygen species are also likely to affect parts of the system other than the substrate, leading to a contamination of the discharge gas. This unwanted contamination, in turn, would adversely affect the quality of the film being produced. The Board accepts the appellant's submissions in this respect and considers that the problem to be solved by the claimed process with regard to D16 is to be seen in the reduction of contamination during the deposition process.

2.3 According to the embodiment of claim 1, the patent in suit proposes to solve the above problem by producing atomic oxygen, ozone or excited oxygen molecules by means of generating a discharge in an oxygen gas or oxygen-containing mixed gas in a pipe through which the gas is supplied.

2.4 The Board notes that the appellant has not submitted any evidence showing that the contamination is indeed

avoided or reduced when the oxygen or oxygen-containing gas is discharged in the gas supply pipe instead of it being discharged outside the pipe as in D16. However, the respondent has not queried the effect of the claimed process. The Board considers it plausible that, by generating the gas discharge in the pipe, contact of the discharge gas with the electrode can be avoided. In this case, contamination is likely to be reduced. The Board therefore accepts that the problem of contamination is solved by the process according to claim 1.

2.5 It remains to be decided whether the proposed solution to the contamination problem is obvious in the light of the available prior art.

The problem of contamination due to electrodes being immersed in an oxygen plasma is addressed in document D17 (page 373, right-hand column, last sentence of second full paragraph). D17 is directed to the application of microwave oxygen discharge in forming thin film oxides on silicon wafers. In particular, it discusses the difference between conventional microwave plasma oxidation and microwave atomic oxygen afterglow oxidation, which is a remote plasma technique. The study reports that, in the conventional system, the plasma is prone to contamination. In contrast, less damage to the growing oxide can be expected when the substrate wafer is located beyond the plasma but within reach of the activated afterglow gas (D17, page 374, left-hand column, first full paragraph).

In order to solve the problem of contamination which may arise in the process of D16, the skilled person would consider the recent literature in the field of

electronics materials relating to controlled oxidation by oxygen plasma. In such a routine search, he could not fail to find D17. He would realise that the solution to the contamination problem given in D17 is applicable to the plasma oxidation process disclosed in D16. In applying the teaching of D17, he would avoid direct contact of discharge gas with the electrode as in D16 and would instead resort to the remote plasma technique for causing the gas discharge. The remote plasma technique as illustrated in Figure 2 of D17 (see page 374: "Experimental Procedure") corresponds to the method of discharging the oxygen or oxygen-containing gas in a pipe as in claim 1.

2.6 The appellant has contended that, in the prior art process according to D17, the object to be oxidised is a formed silicon wafer. Therefore, the skilled person would not turn to D17 for a solution to the present problem which concerns gas contamination during the formation of superconductor thin films.

The Board concedes that D17 is not directed to a process for depositing superconductor thin films. However, the Board holds that D17 is in a technical field closely related to the present application in the sense that both are related to the growth of an oxide thin film on a substrate. In both processes, excited oxygen species are generated by an electrical discharge of an oxygen-containing gas and supplied to the substrate for oxidation.

Moreover, the problem of contamination treated in D17 is entailed by the method of gas discharge. The Board holds that the contamination problem and the solution as discussed in D17 are not inherently different,

whether the product of the oxidation with the discharge gas be an oxide superconductor formed from reactant gases as in the patent in suit or an oxide film obtained by oxidation of the surface of a wafer as in D17.

2.7 For these reasons, the process according to claim 1 does not involve an inventive step within the meaning of Article 56 EPC. The main request is therefore not allowable.

Auxiliary requests 1 to 6

3. Claim 1 of each of the auxiliary requests 1 to 6 has been amended to include the functional feature of "the quantity of atomic oxygen, ozone or excited oxygen molecules supplied being sufficient to form said oxide superconductor thin film".

3.1 As explained by the appellant, this amendment has been made in order to more clearly distinguish the claimed process from the prior art. It is thus undisputed that the feature in question is meant to make a technical contribution to the subject-matter of the claim. It is also not refuted by the appellant that the added feature is not explicitly disclosed in the application as originally filed.

In this case, the Board has to assess whether the skilled person can clearly and unambiguously infer this feature from the original description, taking into account his common general knowledge.

3.2 The application as originally filed is directed to the forming of superconductor thin films with excited

oxygen being supplied to or near a thin film deposition site. It is further specified that the excited oxygen may be one or more of atomic oxygen, ozone, excited oxygen molecule, oxygen molecule ion and oxygen atom ion (see the European patent application published in accordance with Article 158(3) EPC, column 4, lines 18 to 24 and column 3, lines 50 to 53).

Methods for obtaining these oxygen species are recited in the application. In the preferred embodiments such as illustrated in Figure 1, Figure 3 and Figure 5, the methods for exciting the oxygen or oxygen-containing gas involve radio frequency (RF) gas discharge, silent discharge and microwave discharge respectively (see also column 5, line 15; column 6, line 3 and column 6, line 27).

The Board therefore concurs with the appellant insofar as the neutral species (atomic oxygen, ozone or excited oxygen molecules) are among the excited oxygen species recited in the application as originally filed. It is also accepted that various methods of gas discharge disclosed in the application are capable of producing these species. Therefore, the stipulation in claim 1 that these particular species are supplied to or near the deposition site has a basis in the application as filed.

3.3 It is, however, undisputed that ions are also formed during the electric discharge. In fact, oxygen ions are expressly described in the original application as being among the excited oxygen species formed during the electric discharge and susceptible of producing the superconductor thin film (see point 3.2). A difference in behaviour or effect between these oxygen species is

not reported therein. On the contrary, the ions are referred to in exactly the same manner as the other, neutral species.

Moreover, the application does not reveal the relative amount of neutral oxygen species with respect to the amount of ions in the discharge gas. Details of the gas discharge are given only for the single example at column 12, lines 3 to 7 of the description, without, however, indicating which excited oxygen species are actually formed.

3.4 The appellant has asserted that the gas discharge methods mentioned in the application generate atomic oxygen, ozone and excited oxygen molecules in an amount several times larger than that of oxygen ions. Further, even if a small amount of ions is generated in the supply pipe, the ions have recombined with electrons to produce radicals by the time they reach the substrate. Thus, the quantity of ions inherently present in the excited gas produced by the methods of the invention is so small as to be insignificant. In support of this assertion, the appellant has referred to results shown in documents D12, D13 and D15 (see Grounds of Appeal dated 19 March 1998, points 2, 4, 6 and 8; letter dated 10 June 1999, page 1, penultimate paragraph, page 2, paragraphs 4 and 7 and page 3, paragraphs 2 to 4).

The Board notes that the documents referred to by the appellant are reports on very specific studies, namely "Density measurement of O atoms in helicon wave oxygen discharge"(D12), "Ozone synthesis from oxygen" (D13) and "MBE method using radical source" (D15). Furthermore, two of these documents were even published after the filing date of the application (D12,

published July 1994 and D15, published 1992). These citations clearly do not reflect the common general knowledge in the art available to the skilled person at the filing date. Therefore, they cannot be used to interpret the disclosure of the application.

3.5 The appellant has also advanced the argument that ions are recognised as having a detrimental effect on the growth of these films. The person skilled in the art would realise that atomic oxygen, ozone and excited oxygen molecules are entirely responsible for the film quality. Further, the whole tenor of the application is the forming of superconductor thin films. The original application therefore implicitly discloses that atomic oxygen, ozone and excited oxygen molecules are supplied in sufficient quantities, otherwise the superconductor thin films could not be formed.

The appellant has made specific reference to document D16 in support of his above assertion as to the detrimental effect of oxygen ions (see letter of 19 March 1998, point 5; letter of 10 June 1999, page 3, paragraphs 5 and 6). The Board notes that this prior art document discusses a particular method for growing superconductor thin films, which cannot be considered as part of the common general knowledge. Therefore, the specific teaching of D16 cannot be used to interpret the application as originally filed.

On the other hand, there are recent prior art documents disclosing the use of oxygen ions in the preparation of superconducting oxide films, see e.g. D10, claim 2. The alleged detrimental effect of ions, therefore, cannot be considered as common general knowledge. Thus, there is no evidence on file that superconductor thin films

could not be formed without excited neutral oxygen species.

3.6 In the context of the patent in suit, the added functional feature of supplying atomic oxygen, ozone or excited oxygen molecules in sufficient quantity to form said oxide superconductor thin film implies that the formation of the ionic species is suppressed to the extent that the quantity of the neutral species alone is sufficient to form said oxide thin film.

From the above discussion, the Board finds that the skilled person cannot clearly and unambiguously deduce from the original application that essentially the excited neutral oxygen species and not oxygen ions are responsible for producing the superconducting film. It follows that a feature having this implication is not properly based on the application as originally filed. Consequently, the incorporation of this functional feature into the claim constitutes added subject-matter. Amended claims comprising this functional feature therefore do not fulfill the requirement of Article 123(2) EPC.

Auxiliary requests 1 to 6 are not allowable because claim 1 of each of these requests comprises said unallowable functional feature.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:	The Chairman:
S. Hue	G. Wassenaar