BESCHWERDEKAMMERN DES EUROPÄISCHEN PATENTAMTS

BOARDS OF APPEAL OF THE EUROPEAN PATENT OFFICE

CHAMBRES DE RECOURS DE L'OFFICE EUROPEEN DES BREVETS

A		B		C	X

File No.: T 0216/91 - 3.4.1

Application No.: 87 301 834.5

Publication No.: 0 237 250

Classification: H01L 31/08

Title of invention: Ferroelectric film device and process for producing the same

D E C I S I O N
of 14 July 1993

Applicant: MATSUSHITA ELECTRIC INDUSTRIAL CO., LTD.

Proprietor of the patent:

Opponent:

Headword:

EPC:

Keyword: Inventive step (yes), after clarifying that the effects to be produced are an inherent property of a device feature defined via a quantitatively measurable parameter.
Decisions cited: T 94/82, OJ EPO 1984, 75

Headnote
Catchwords

Europäisches Patentamt | European Patent Office | Office européen des brevets

Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number: T 0216/91 - 3.4.1

D E C I S I O N
of the Technical Board of Appeal 3.4.1
of 14 July 1993

Appellant: MATSUSHITA ELECTRIC INDUSTRIAL CO., LTD.
1006, Oaza Kadoma
Kadoma-shi,
Osaka-fu, 571
JAPON

Representative: Ablett, Graham Keith
F.J. Cleveland & Company
40-43 Chancery Lane
London WC2A 1 JQ
GRANDE BRETAGNE

Decision under appeal: **Decision of the Examining Division 048 of the European Patent Office dated 17 October 1990 refusing European patent application No. 87 301 834.5 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman: G.D. Paterson
Members: H.J. Reich
U.G.O.M. Himmler

Summary of Facts and Submissions

I. European patent application No. 87 301 834.5 (publication No. 0 237 250) was refused by decision of the Examining Division.

II. The reason given for the refusal was that the subject-matter of Claim 1 as filed on 18 July 1990 - comprising *inter alia* the wording:

"and in that at least 75% of the polarisation axes in the thin film are oriented in substantially perpendicular to the thin film surface, the spontaneous polarisation of the domains in the film having been achieved without need for poling treatment" -

did not satisfy the requirements of Articles 52 and 56 EPC having regard to documents:

D2: DE-B2-2 061 447,
D3: GB-A-1 516 408,
D4: Journal of Crystal Growth; Vol. 45, December 1978, pages 393 to 398, and
D6: Maissel and Glang: "Handbook of Thin Film Technology", MacGraw-Hill, 1970, pages 15-10 to 15-14.

The Examination Division in essence took the following view: Compositions according to the claimed chemical formula $(Pb_x\ La_y)(Ti_z\ Zr_w)O_3$ (PLZT) are known e.g. from documents D2 and D3. The selection of the claimed ranges for x, y, z and w must be regarded as obvious. A **spontaneous** polarisation of PLZT-films appearing without poling treatment is known from document D4. Orientations of the polarisation axes normal to the film plane are known from document D6 to depend on a variety of

deposition parameters. Hence, a deposition parameter selection for a high degree of such orientation would be obvious and its lower value of 75% arbitrary. Due to the fact that the end product does not show whether the claimed degree of polarisation orientation was achieved with or without poling treatment, such process feature has to be disregarded in the evaluation of inventive step.

III. The Appellant lodged an appeal against this decision. In support of an inventive step underlying the subject-matter of Claim 1, the Appellant argued mainly that in the cited prior art, in particular in document D4 there is no suggestion of a PLZT device which has spontaneous polarisation orientated in a single direction without the need for a poling treatment. The wording in document D4, page 393, column 1, line 27: "Semiconductor substrates ... were also used to investigate the possibilities of electronic applications utilising spontaneous polarisation of the ferroelectric PLZT films" cannot be interpreted as a hint to a thin PLZT film in which the polarisation axes of the spontaneous polarisation within **all** domains of the film are orientated in the **same** direction. In the generally known state of the art deposited ferroelectric material adapts a domain structure with no particular correlation between the directions of orientation of spontaneous polarisation in each of the domains. In conventional ferroelectric films only occasionally the statistical distribution of the polarisation directions of the polarisation axes of the domains results in a weak resultant spontaneous polarisation and needs a poling treatment in order to transform the deposited thin film into an unstable, electrostatistically higher energy state wherein the spontaneous polarisation axes of all domains are aligned with each other to one direction. The Appellant made the surprising discovery that certain PLZT thin films having

a defined composition possess perpendicular to their surface self-aligned polarisation axes of their domains without poling treatment, a novel concept, contrary to what had been considered to be the situation according to generally accepted physical laws.

IV. In response to communications of the Board of Appeal expressing *inter alia* doubts whether the subject-matter of Claim 1 would comprise structural device features indicating the omission or application of a poling treatment and inviting to clarify in Claim 1, that the amount of unidirectional polarisation appearing in the thin film without poling treatment - as disclosed in the description - corresponds to the crystallographic c-axis orientation degree α, representing a parameter which is related to the physical structure of the claimed film device and can be determined by an objective procedure, the Appellant now requests that the decision under appeal be set aside and that a patent be granted on the basis of the following documents:

Claims: 1, filed 24 March 1993 and amended 8 June 1993;
2 and 3, filed 5 October 1992;

Description: EP-A1-0 237 250, page 2, lines 1 to 34 until "none of these"; page 2, line 49 from "dependence" to page 6, line 15, with the amendment on page 3, line 8, as requested on 8 June 1993;
pages 3, 3a filed 24 March 1993 and amended 8 June 1993;
page 3b, filed 24 March 1993;

Drawings: Figures 1 to 6 according to EP-A1-0 237 250;
Figures 7 to 9 filed 18 July 1990.

V. Claim 1 reads as follows:

"1. A ferroelectric thin film device comprising a substrate (1), a ferroelectric thin film (3) formed over the substrate, said thin film composition being represented by the chemical formula $(Pb_xLa_y)(Ti_zZr_w)O_3$, and electrodes attached to said ferroelectric film characterized in that the substrate is MgO, the constituents of said film lie in a range selected from

(a) $0.70 \leq x \leq 1$, $0.90 \leq x + y \leq 1$,
$0.95 \leq z \leq 1$, $w = 0$,

(b) $x = 1$, $y = 0$, $0.45 \leq z < 1$, $z + w = 1$, and

(c) $0.83 \leq x \leq 1$, $x + y = 1$, $0.5 \leq z < 1$,
$0.95 \leq z + w \leq 1$,

and in that perpendicular to the thin film surface the crystallographic c-axis orientation degree α is at least 75%, so that at least 75% of the polarization axes in the thin film are orientated along the c-axis and the spontaneous polarization of the domains in the film are orientated in substantially one direction without the need for any poling treatment."

Claims 2 and 3 are dependant on Claim 1.

VI. In support of his request the Appellant argued essentially that in PLZT-type ferroelectric thin films known prior to the priority date of the present application, the polarisation axes were orientated along the crystallographic a-axis (100 plane). Thus, the prior art references do not direct the skilled person to preparing devices having the orientation along the c-axis, allowing a use in an as-grown state, and do not suggest that PLZT thin films with the claimed compositions and grown on an MgO substrate result in the desired film characteristics, i.e. a **self** alignment of the spontaneous polarisation direction within the domains

into one only direction perpendicular to the film surface.

Reasons for the Decision

1. Claim 1 comprises the subject-matter of original Claims 1 and 3 and features derived from the original description, page 4, line 26 to page 6, line 8. Claims 2 and 3 correspond to original Claims 2 and 4. The amendments of the description are in line with Rules 27(1)(b) and (c) EPC. There is, therefore, no objection under Article 123(2) EPC to the current set of application documents.

2. The claimed crystallographic c-axis orientation degree α is defined in the original description page 6, lines 5 to 8, by measurable intensities of X-rays reflected from the 001- and 100-crystal planes respectively and thus represents a parameter which can be reliably determined by an objective experimental procedure in the sense of decision T 94/82, OJ EPO 1984, 75. The c-axis orientation degree α thus unambiguously characterizes the physical structure in the ferroelectric film of the claimed film device in form of a device feature, making any further reference to a process step superfluous. In the Board's view, the additional wording of Claim 1 concerning the orientation of the polarisation axes (i.e. the direction of the spontaneous polarisation of the domains) in one direction along the c-axis without poling treatment only explains the fact that such a polarisation is an inherent property of the crystallographic c-axis orientation perpendicular to the film surface in the film as grown. For the above reasons Claim 1 satisfies Article 84 EPC.

3. Novelty

3.1 The prior art disclosed in document D4 or in ESR-document

D5: Japanese Journal of Applied Physics, Vol. 16, 1977, No. 9, pages 1707 and 1708

comprises only the features in the first part of Claim 1. Due to the fact that in Figures 1 of documents D4 and D5 no 001 X-ray reflection peak was unambiguously recorded, these conventional thin film devices can be regarded to have perpendicular to the film surface no c-axis orientation. In the Board's view, the recorded intensity at the 011 angle in Figure 1 of document D5 falls within the noise level and is of negligible intensity.

3.2 Documents D2, D3 and the remaining ESR-documents concern no thin films but pressed ceramic plates.

3.3 Thus, the subject-matte of Claim 1 is considered novel in the sense of Article 54 EPC.

4. Inventive step

4.1 Starting from the closest prior art device according to document D4, the objective problem underlying the present invention is to provide a ferroelectric thin film device which allows to take out an output caused by changes of the spontaneous polarisation at the optimal condition of a **unidirectional** spontaneous polarisation without previous poling treatment of the film (i.e. not making use of the ferroelectric remanence effect); see the published description page 2, lines 15 to 17 and 39, 40.

4.2 This problem is solved by the device as claimed having a ferroelectric thin film with a crystallographic c-axis orientation perpendicular to the film surface, the

experimentally measurable degree α of which is at least 75%.

4.3 In the Board's view, the wording "spontaneous polarization" in document D4, page 393, right-hand column, line 1, is not interpreted by a skilled person as a **self-aligned unidirectional** spontaneous polarisation. Moreover, there is no hint in document D4 or D5, that the volume of grown domains with the same direction of electrical polarisation depends on the crystallographic orientation, in which the film is grown. The Appellant is followed in his view, that the X-ray diffraction patterns of the thin film devices of documents D4 and D5 are to be interpreted in that in these known devices the crystallographic **a-axis** is orientated perpendicular to the film axis (and thus the c-axis parallel to the film surface). In the Board's opinion, a skilled person - on the basis of his general knowledge - is not able to foresee that a unidirectional spontaneous polarisation (i.e. a film device forming in total a region of one domain) can be realized by allowing the film to grow with the crystallographic **c-axis** perpendicular to its surface.

4.4 The composition ratios of the PLZT material disclosed in documents D2 and D3 are chosen in order to optimize the optical birefringeance of a ceramic plate. In the Board's view, from such a technical teaching no logical conclusions can be drawn with regard to a PLZT material composition appropriate for a unidirectional spontaneous electrical polarization of such material when deposited in form of a thin film.

4.5 Document D1 relates to a different material. In the Board's view, the fact that a certain degree of unipolarity was observed in Al-Ba-Sr-TiO_3-Si-layers does not allow to expect unipolarity in a Pb-La-Ti-Zr-O_3-layer or to relate this effect to a particular crystallographic

orientation of the layer. Document D6 only mentions a c-axis orientation perpendicular to a CdS film surface without informing about the electrical polarization of the CdS film.

4.6 For the reasons set out in paragraphs 4.1 to 4.5, the subject-matter of Claim 1 is considered to involve an inventive step in the sense of Article 56 EPC.

5. Thus, Claim 1 is allowable under Article 52(1) EPC. Dependent Claims 2 and 3 concern particular embodiments of the device claimed in Claim 1 and are, therefore, likewise allowable.

Order

For these reasons, it is decided that:

1. The decision of the Examining Division is set aside.

2. The case is remitted to the first instance in order to grant a patent on the basis of the following documents:

Claims: 1, filed 24 March 1993 and amended 8 June 1993;
2 and 3, filed 5 October 1992;

Description: EP-A1-0 237 250, page 2, lines 1 to 34 (until "none of these"); page 2, line 49 (from "dependence") to page 6, line 15, and amended 8 June 1993;
pages 3, 3a, 3b filed 24 March 1993 and amended 8 June 1993;

Drawings: Figures 1 to 6 according to EP-A1-0 237 250;
Figures 7 to 9 filed 18 July 1990.

The Registrar: The Chairman:

M. Beer G.D. Paterson