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### COMPOSITE SUBSTRATE

#### Abstract

A composite substrate including: a first piezoelectric layer; a second piezoelectric layer that is disposed to be stacked on the first piezoelectric layer; and a support substrate that supports the first piezoelectric layer and the second piezoelectric layer, an amorphous layer being formed on a bonding interface between at least one of the first piezoelectric layer and the second piezoelectric layer and another layer, the second piezoelectric layer, the first piezoelectric layer, and the support substrate being stacked in this order.

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## Background/Summary

CROSS-REFERENCE TO RELATED APPLICATION [0001] This application is a continuation application of PCT/JP2023/041547, filed on Nov. 17, 2023, which claims the benefit of priority of Japanese Patent Application No. 2022-185043, filed on Nov. 18, 2022, the entire contents of which are incorporated herein by reference.

### 1. TECHNICAL FIELD

[0002] The present invention relates to a composite substrate.

### 2. DESCRIPTION OF RELATED ART

[0003] In the related art, piezoelectric actuators that cause electromechanical conversion films to vibrate have been put into practical use in liquid droplet ejection heads of inkjet recording apparatuses and the like. Furthermore, applications to other purposes (for example, MEMS mirror devices for head-up displays) have also been expected in recent years.

[0004] As a method of manufacturing a piezoelectric actuator in the related art, a technique of producing a composite substrate in which a piezoelectric element is caused to adhere to a silicon substrate via a metal layer and configuring a piezoelectric actuator using the composite substrate is known (Patent Literature 1: Japanese Patent Laid-Open No. 2007-271788). Moreover, a technique of producing a composite substrate in which a piezoelectric layer of PZT or the like is formed on a silicon substrate by a sputtering method and configuring a piezoelectric actuator using the composite substrate is also known (Patent Literature 2: Japanese Patent Laid-Open No. 2014-225596).

[0005] In the composite substrate produced using the method disclosed in Patent Literature 1, the piezoelectric element cannot be formed to be very thin in order to enable the piezoelectric element to adhere to the silicon substrate. Therefore, there is a problem that it is difficult to achieve size reduction of the piezoelectric actuator. On the other hand, the composite substrate produced using the method disclosed in Patent Literature 2 is advantageous for size reduction of the piezoelectric actuator since the piezoelectric layer can be formed to be thin in the film formation. However, since the film formation is performed in a high-temperature environment, deformation such as warpage is likely to occur in the composite substrate due to a change in temperature after the film formation. Therefore, there is a problem that it is difficult to increase the thickness of the piezoelectric layer.

[0006] In a case where the amount of operation required for a piezoelectric layer is relatively large as in a piezoelectric actuator of a MEMS mirror device, for example, it is necessary to achieve both size reduction and a piezoelectric property. However, it is difficult to realize this by the methods in Patent Literatures 1 and 2.

[0007] The present invention was made in view of the above circumstances, and a main object thereof is to provide a composite substrate that includes piezoelectric layers capable of achieving both size reduction and a piezoelectric property.

### SUMMARY OF THE INVENTION

[0008] A composite substrate according to a first aspect of the present invention includes: a first piezoelectric layer; and a second piezoelectric layer that is disposed to be stacked on the first piezoelectric layer, and an amorphous layer is formed on a bonding interface between at least one of the first piezoelectric layer and the second piezoelectric layer and another layer.

[0009] A composite substrate according to a second aspect of the present invention includes: a first piezoelectric layer; and a second piezoelectric layer that is disposed to be stacked on the first piezoelectric layer, and at least one of the first piezoelectric layer and the second piezoelectric layer

is directly bonded to another layer.

[0010] According to the present invention, it is possible to provide a composite substrate that includes piezoelectric layers capable of achieving both size reduction and a piezoelectric property.

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## Description

### BRIEF DESCRIPTION OF THE DRAWINGS

[0011] FIG. 1 is a schematic sectional view illustrating an overview configuration of a composite substrate according to a first embodiment of the present invention.

[0012] FIG. 2 is a schematic sectional view illustrating an overview configuration of a composite substrate according to a second embodiment of the present invention.

[0013] FIG. 3 is a schematic sectional view illustrating an overview configuration of a composite substrate according to a third embodiment of the present invention.

[0014] FIG. 4 is a schematic sectional view illustrating an overview configuration of a composite substrate according to a fourth embodiment of the present invention.

[0015] FIG. 5 is a schematic sectional view illustrating an overview configuration of a composite substrate according to a fifth embodiment of the present invention.

[0016] FIG. 6 is a diagram illustrating an example of a case where an opening portion is provided in a support substrate in the composite substrate according to the fifth embodiment of the present invention.

[0017] FIG. 7 is a diagram illustrating an example of a case where the opening portion is provided in the support substrate in the composite substrate according to the fifth embodiment of the present invention.

[0018] FIG. 8 is a diagram illustrating an example of a case where a hollow portion is provided in the support substrate in the composite substrate according to the fifth embodiment of the present invention.

[0019] FIG. 9 is a diagram illustrating an example of a case where a sacrificial layer is provided in the support substrate in the composite substrate according to the fifth embodiment of the present invention.

[0020] FIG. 10 is a schematic sectional view illustrating an overview configuration of a composite substrate according to a sixth embodiment of the present invention.

[0021] FIGS. 11A, 11B and 11C are diagrams illustrating examples of manufacturing processes of the composite substrate according to the second embodiment of the present invention.

[0022] FIG. 12A is a sectional TEM observation photograph (50,000 folds) of a composite substrate in an example.

[0023] FIG. 12B is a sectional TEM observation photograph (400,000 folds) of the composite substrate in the example.

[0024] FIG. 12C is a sectional TEM observation photograph (2,000,000 folds) of the composite substrate in the example.

### DESCRIPTION OF PREFERRED EMBODIMENTS

[0025] Although embodiments of the present invention will be described below with reference to the drawings, the present invention is not limited to these embodiments. Also, although the width, the thickness, the shape, and the like of each component may be schematically represented in the drawings as compared with those in the embodiments for more clear explanation, these are just examples and are not intended to limit interpretation of the present invention.

#### A. Composite Substrate

##### (A-1) First Embodiment

[0026] FIG. 1 is a schematic sectional view illustrating an overview configuration of a composite substrate according to a first embodiment of the present invention. A composite substrate 100

includes a first piezoelectric layer **10** and a second piezoelectric layer **20** that is disposed to be stacked on the first piezoelectric layer **10**. Polarization directions of the first piezoelectric layer **10** and the second piezoelectric layer **20** are preferably mutually opposite directions, and these forms a bimorph structure.

[0027] In the composite substrate **100**, the first piezoelectric layer **10** and the second piezoelectric layer **20** are directly bonded to each other, and an amorphous layer generated at the time of the bonding is formed at a bonding interface therebetween. Note that specific examples and a bonding method of the first piezoelectric layer **10** and the second piezoelectric layer **20** will be described later.

#### (A-2) Second Embodiment

[0028] FIG. **2** is a schematic sectional view illustrating an overview configuration of a composite substrate according to a second embodiment of the present invention. A composite substrate **110** further includes an electrode layer **30** disposed between a first piezoelectric layer **10** and a second piezoelectric layer **20** as compared with the composite substrate **100** described in the first embodiment. The electrode layer **30** is constituted using a conductive material such as metal, for example.

[0029] In the composite substrate **110**, the first piezoelectric layer **10** or the second piezoelectric layer **20** and the electrode layer **30** are directly bonded to each other, and an amorphous layer generated at the time of the bonding is formed at a bonding interface therebetween. Note that a specific example and a bonding method of the electrode layer **30** will be described later.

#### (A-3) Third Embodiment

[0030] FIG. **3** is a schematic sectional view illustrating an overview configuration of a composite substrate according to a third embodiment of the present invention. A composite substrate **120** includes a bonding layer **40**, instead of the electrode layer **30**, disposed between a first piezoelectric layer **10** and a second piezoelectric layer **20** as compared with the composite substrate **110** described in the second embodiment. The bonding layer **40** is constituted using an amorphous body, for example.

[0031] In the composite substrate **120**, the first piezoelectric layer **10** or the second piezoelectric layer **20** and the bonding layer **40** are directly bonded to each other, and an amorphous layer generated at the time of the bonding is formed at a bonding interface therebetween. Note that a specific example and a bonding method of the bonding layer **40** will be described later.

#### (A-4) Fourth Embodiment

[0032] FIG. **4** is a schematic sectional view illustrating an overview configuration of a composite substrate according to a fourth embodiment of the present invention. A composite substrate **130** further includes a support substrate **50** that supports a first piezoelectric layer **10** and a second piezoelectric layer **20** as compared with the composite substrate **100** described in the first embodiment. The support substrate **50** is constituted using an arbitrary material.

[0033] In the composite substrate **130**, the first piezoelectric layer **10** and the second piezoelectric layer **20** are directly bonded to each other, and an amorphous layer generated at the time of the bonding is formed at a bonding interface therebetween. Similarly, the first piezoelectric layer **10** and the support substrate **50** are directly bonded to each other, and an amorphous layer generated at the time of the bonding is formed at a bonding interface therebetween. Note that a specific example and a bonding method of the support substrate **50** will be described later.

#### (A-5) Fifth Embodiment

[0034] FIG. **5** is a schematic sectional view illustrating an overview configuration of a composite substrate according to a fifth embodiment of the present invention. A composite substrate **140** further includes an electrode layer **31** disposed between a support substrate **50** and a first piezoelectric layer **10** and an electrode layer **32** disposed between the first piezoelectric layer **10** and a second piezoelectric layer **20** as compared with the composite substrate **130** described in the fourth embodiment. The electrode layers **31** and **32** are constituted using a conductive material

such as metal, for example, similarly to the electrode layer **30** in the composite substrate **110** described in the second embodiment. Note that the support substrate **50** may have a structure as in FIGS. **6**, **7**, **8**, and **9** in the composite substrate **140**. FIGS. **6** and **7** illustrate an example of a case where an opening portion is provided in the support substrate **50** in the composite substrate **140** according to the fifth embodiment of the present invention. FIG. **8** illustrates an example of a case where a hollow portion is provided in the support substrate **50** in the composite substrate **140** according to the fifth embodiment of the present invention. FIG. **9** illustrates an example of a case where a sacrificial layer to be removed in a later process is provided in the support substrate **50** in the composite substrate **140** according to the fifth embodiment of the present invention.

[0035] In the composite substrate **140**, the support substrate **50** or the first piezoelectric layer **10** and the electrode layer **31** are directly bonded to each other, the first piezoelectric layer **10** or the second piezoelectric layer **20** and the electrode layer **32** are directly bonded to each other, and amorphous layers generated at the time of the bonding are formed at the bonding interfaces therebetween. Note that specific examples and bonding methods of the electrode layers **31** and **32** will be described later.

#### (A-6) Sixth Embodiment

[0036] FIG. **10** is a schematic sectional view illustrating an overview configuration of a composite substrate according to a sixth embodiment of the present invention. A composite substrate **150** includes piezoelectric layers **11** and **12** that have a common polarization direction instead a first piezoelectric layer **10** and a second piezoelectric layer **20** and further includes an electrode layer **33** that is formed on a surface of the piezoelectric layer **12** as compared with the composite substrate **140** described in the fifth embodiment. The electrode layer **33** is constituted using a conductive material such as metal, for example, similarly to the electrode layer **30** in the composite substrate **110** described in the second embodiment.

[0037] In the composite substrate **150**, a support substrate **50** or the piezoelectric layer **11** and an electrode layer **31** are directly bonded to each other and the piezoelectric layer **11** or the piezoelectric layer **12** and an electrode layer **32** are directly bonded to each other similarly to the composite substrate **140** described in the fifth embodiment, and amorphous layers generated at the time of the bonding are formed at bonding interfaces therebetween.

[0038] The composite substrates **100** to **150** described above in the embodiments are used as piezoelectric actuators which are, for example, MEMS devices. Note that although not illustrated in the drawings, the composite substrates **100** to **150** may further include arbitrary layers. Types, functions, numbers, combinations, arrangement, and the like of such layers may be appropriately set in accordance with purposes. For example, the composite substrates **100** to **140** may further include an electrode layer **33** that is disposed on the second piezoelectric layer **20** similarly to the composite substrate **150**. The electrode layer **33** is provided with, for example, a wiring layer. Also, an electrode may be formed on an exposed surface of the first piezoelectric layer **10** in the composite substrates **100** to **120**, or an electrode may be formed on an exposed surface of the first piezoelectric layer **10** formed by removing the support substrate **50** through etching or the like in the composite substrate **130**, for example.

[0039] The composite substrates **100** to **150** may be manufactured into arbitrary appropriate shapes. In one embodiment, the composite substrates **100** to **150** may be manufactured in the form of so-called wafers. Also, the sizes of the composite substrates **100** to **150** may be appropriately set to wafer (substrate) diameters of 50 mm to 150 mm, for example, in accordance with purposes.

#### A-1. Piezoelectric Layer

[0040] The first piezoelectric layer **10** and the second piezoelectric layer **20** (hereinafter, simply referred to as “piezoelectric layers”) are constituted by non-oriented polycrystalline bodies, for example. Also, as a representative example, the piezoelectric layers are constituted by sintered bodies. For example, grain boundaries are observed in the piezoelectric layers in TEM observation. It is possible to obtain the composite substrates **100** to **150** that include piezoelectric layers capable

of achieving both size reduction and a piezoelectric property by employing such configurations. Specifically, since the piezoelectric layers may be formed alone, an internal stress does not occur due to interactions with other members at the time of the formation of the piezoelectric layers, for example, and it is thus possible to suppress deformation such as warpage. Also, options for a material constituting the piezoelectric layers increases by constituting the piezoelectric layers using non-oriented polycrystalline bodies, and it is possible to address diversified properties. Specifically, it is possible to finely adjust properties such as piezoelectric constants, dielectric constants, electromechanical coupling coefficients, and curie temperatures in accordance with needs. Furthermore, it is possible to form the piezoelectric layers at low costs, which may contribute to an improvement in reliability of the obtained composite substrates **100 to 150**.

[0041] Here, “non-oriented” means that a c-axis orientation degree obtained by the Lotgering method is 80% or less, is preferably 60% or less, is more preferably 40% or less, is further preferably 20% or less, and is particularly preferably 10% or less. The c-axis orientation degree is an orientation degree  $F_{(001)}$  of a (001) plane calculated using the following expressions from an XRD profile obtained by measurement using an X-ray diffraction apparatus.

$$[00001] F_{(001)} = (p - p_0) / (1 - p_0) \times 100 \quad p = \text{Math. } I(001) / \text{Math. } I(hk1)$$

$$p_0 = \text{Math. } I_0(001) / \text{Math. } I_0(hk1)$$

( $I$  and  $I_{\text{sub}0}$  represent diffraction intensity, and  $p$  and  $p_0$  are calculated from a ratio between diffraction intensity derived from a c-axis diffraction plane (001) and diffraction intensity of a total diffraction plane (hk1);  $I$  and  $p$  are values obtained from an XRD profile of a piezoelectric film (piezoelectric substrate), and  $I_{\text{sub}0}$  and  $p_0$  are values obtained from an XRD profile of a sample obtained by forming the piezoelectric film (piezoelectric substrate) into powder.)

[0042] As a material constituting the piezoelectric layer, an arbitrary appropriate ferroelectric body is used. A PZT (lead zirconate titanate)-based compound is preferably used. As the PZT-based compound, it is possible to use not only a two-component-based PZT (PbZrO<sub>3</sub>-PbTiO<sub>3</sub>) of lead titanate having a perovskite structure and lead zirconate but also a three-component-based PZT. The piezoelectric layers can contain the three-component-based PZT by constituting the piezoelectric layers by non-oriented polycrystalline bodies. It is possible to cause the obtained composite substrates **100 to 150** to address diversified properties by using the three-component-based PZT. Specifically, it is possible to finely adjust properties such as piezoelectric constants, dielectric constants, electromechanical coupling coefficients, and curie temperatures in accordance with needs.

[0043] An atomic ratio (Zr/Ti) between Zr and Ti contained in the piezoelectric layers is preferably 0.7 or more and 2.0 or less, and is more preferably 0.9 or more and 1.5 or less.

[0044] The three-component-based PZT is representatively expressed as ATiO<sub>3</sub>-PbZrO<sub>3</sub>-PbTiO<sub>3</sub> or PbBO<sub>3</sub>-PbZrO<sub>3</sub>-PbTiO<sub>3</sub>, where A and B each represent an element other than Pb, Zr, and Ti. Examples of the element A contained in the third component in the three-component-based PZT include Li, Na, K, Bi, La, Ce, and Nd. Examples of the element B contained in the third component of the three-component-based PZT include Li, Cu, Mg, Ni, Zn, Mn, Co, Sn, Fe, Cd, Sb, Al, Yb, In, Sc, Y, Nb, Ta, Bi, W, Te, and Re. One of these may be used alone, or two or more kinds thereof may be used in combination.

[0045] A proportion of the third component with respect to a total of Zr, Ti, Pb, and the third component (the element A and/or the element B) contained in the piezoelectric layers, specifically, the atomic ratio of the third component/(Zr+Ti+Pb+ the third component) is preferably 0.05 or more and 0.25 or less, and is more preferably 0.10 or more and 0.20 or less.

[0046] The atomic ratio (proportion) can be obtained by composition analysis based on energy dispersive X-ray spectroscopy (EDX).

[0047] Other specific examples of the material constituting the piezoelectric layers include PMN-PT (Pb(Mg<sub>1/3</sub>Nb<sub>2/3</sub>)O<sub>3</sub>-PbTiO<sub>3</sub>), barium titanate (BaTiO<sub>3</sub>), lead titanate (PbTiO<sub>3</sub>), lead

metaniobate ( $\text{PbNb}_2\text{O}_6$ ), bismuth titanate ( $\text{Bi}_4\text{Ti}_3\text{O}_{12}$ ), KNN( $(\text{K}_{0.5}\text{Na}_{0.5})\text{NbO}_3$ ), KNN-LN( $((\text{K}_{0.5}\text{Na}_{0.5})\text{NbO}_3)\text{—LiNbO}_3$ ), and BT-BNT-BKT ( $(\text{Bi}_{0.5}\text{Na}_{0.5})\text{TiO}_3\text{—}(\text{Bi}_{0.5}\text{K}_{0.5})\text{TiO}_3\text{—BaTiO}_3$ ).

[0048] The piezoelectric layers may be any piezoelectric layers as long as they can be formed alone, and may be monocrystal bodies. In the case of monocrystal bodies, the piezoelectric layers are produced by slicing a monocrystal ingot. Specific examples of the monocrystal bodies include  $\text{LiTaO}_3$ ,  $\text{LiNbO}_3$ , and crystal.

[0049] The thickness of the piezoelectric layers is, for example, greater than  $0.2\text{ }\mu\text{m}$ , is preferably  $0.3\text{ }\mu\text{m}$  or more, is more preferably  $1\text{ }\mu\text{m}$  or more, and is further preferably  $3\text{ }\mu\text{m}$  or more. In one embodiment, the thickness of the piezoelectric layers may be  $5\text{ }\mu\text{m}$  or more, or may be  $6\text{ }\mu\text{m}$  or more. With such a thickness, it is possible to obtain an actuator with large displacement driven at a low voltage, for example. In a case where the piezoelectric layers are formed through film formation such as sputtering as in the related art, for example, it is difficult to achieve such a thickness due to relationships of a film stress, productivity, and the like of the obtained piezoelectric layers. On the other hand, such a thickness can be set by constituting the piezoelectric layers by the non-oriented polycrystalline bodies as described above. Moreover, it is possible to obtain the composite substrates **100** to **150** that reduce occurrence of warpage and the like even with such a thickness by constituting the piezoelectric layers by the non-oriented polycrystalline bodies. On the other hand, the thickness of the piezoelectric layers is, for example,  $200\text{ }\mu\text{m}$  or less, is preferably  $150\text{ }\mu\text{m}$  or less, is more preferably  $100\text{ }\mu\text{m}$  or less, is further preferably  $50\text{ }\mu\text{m}$  or less, and is particularly preferably  $20\text{ }\mu\text{m}$  or less. With such a thickness, it is possible to suppress disadvantages (for example, occurrence of cracking due to heating) occurring due to a difference in thermal expansion from the support substrate and to address a heating process (at  $100^\circ\text{C}$ . or higher, for example) in production of a piezoelectric device, for example. Specifically, it is possible to address mask formation using photolithography in production of a MEMS device.

[0050] As described above, the piezoelectric layers may be constituted by sintered bodies. The sintered bodies may be formed by an arbitrary appropriate method. In one embodiment, the sintered bodies can be formed by pressure-sintering ingredient powder. In a specific example, it is possible to form the sintered bodies by pressure-sintering ingredient powder mixed at a predetermined blending ratio or powder obtained by pulverizing ingredient powder mixed at a predetermined blending ratio into a predetermined particle diameter (for example,  $0.1\text{ }\mu\text{m}$  to  $10\text{ }\mu\text{m}$ ) after calcination. As the pressure-sintering, an arbitrary appropriate method may be employed. Specifically, an HIP method, a hot-press method, or the like may be employed.

[0051] The piezoelectric layers can be obtained by performing working such as grinding and polishing on the sintered bodies (piezoelectric substrates) into a desired thickness, for example. In the formation of the piezoelectric layers, polarization processing is performed at an arbitrary appropriate timing. In one embodiment, a pair of electrodes are provided on mutually facing surfaces of the sintered bodies (piezoelectric substrates) formed into a plate shape, polarization processing is performed with an electric field in a direction from one electrode to the other electrode, and working such as grinding and polishing is performed thereon, thereby obtaining the piezoelectric layers.

[0052] Arithmetic mean roughness  $R_a$  of the piezoelectric layers after the polishing is preferably  $2\text{ nm}$  or less, is more preferably  $1\text{ nm}$  or less, and is further preferably  $0.3\text{ nm}$  or less.

## A-2. Support Substrate

[0053] As the support substrate **50**, an arbitrary appropriate substrate may be used. The support substrate **50** may be constituted by a monocrystal body or may be constituted by a polycrystalline body. Alternatively, the support substrate **50** may be constituted by metal. A material constituting the support substrate **50** is preferably selected from the group consisting of silicon, sialon, sapphire, cordierite, mullite, glass, quartz, crystal, alumina, stainless steel, an iron-nickel alloy (Alloy 42), and brass.

[0054] Silicon may be monocrystal silicon, polycrystalline silicon, or high-resistance silicon. Also, the support substrate **50** may be a silicon-on-insulator (SOI).

[0055] Sialon is representatively a ceramic obtained by sintering a mixture of silicon nitride and alumina and has, for example, a composition represented as  $\text{Si}_6\text{-wAl}_w\text{O}_w\text{N}_8\text{-w}$ . Specifically, sialon has a composition in which alumina is mixed in silicon nitride, and w in the formula denotes a mixing ratio of alumina. Preferably, w is 0.5 or more and 4.0 or less.

[0056] Sapphire is representatively a monocrystal body that has a composition of  $\text{Al}_2\text{O}_3$ , and alumina is a polycrystalline body that has a composition of  $\text{Al}_2\text{O}_3$ . Alumina is preferably light transmitting alumina.

[0057] Cordierite is representatively a ceramic that has a composition of  $2\text{Mg}_0.2\text{Al}_2\text{O}_3.5\text{SiO}_2$ , and mullite is a ceramic that has a composition within a range from  $3\text{Al}_2\text{O}_3.2\text{SiO}_2$  to  $2\text{Al}_2\text{O}_3.\text{Math}.\text{SiO}_2$ .

[0058] In a case of utilization as a piezoelectric actuator, it is also possible to use a support substrate provided with an opening portion or a hollow portion. Examples are illustrated in FIGS. **6**, **7**, and **8**. Moreover, it is also possible to use a support substrate provided with a sacrificial layer. An example is illustrated in FIG. **9**. It is possible to reduce fracture failures at the time of production of the composite substrate by removing the sacrificial layer through etching after formation of the composite substrate. As the sacrificial layer, an arbitrary appropriate configuration may be employed in accordance with purposes. Examples of a material constituting the sacrificial layer include amorphous silicon, silicon, molybdenum, silicon oxide, aluminum oxide, a compound of these materials, and a mixture of these materials. Also, examples of a method of film formation for the sacrificial layer include sputtering, plating, and deposition. Examples of a method of etching the sacrificial layer include wet etching and dry etching.

[0059] As the thickness of the support substrate **50**, an arbitrary appropriate thickness may be employed. The thickness of the support substrate **50** is, for example, 100  $\mu\text{m}$  to 1000  $\mu\text{m}$ .

### A-3. Bonding Layer

[0060] Examples of a material constituting the bonding layer **40** include silicon, tantalum oxide, niobium oxide, aluminum oxide, titanium oxide, and hafnium oxide. The thickness of the bonding layer **40** is, for example, 5 nm to 1  $\mu\text{m}$  and is preferably 10 nm to 200 nm.

[0061] The bonding layer **40** is representatively constituted by an amorphous body. Specifically, the bonding layer **40** may be an amorphous layer. Constituting the bonding layer **40** by an amorphous body makes it easy to perform polishing, which will be described later, for example, and to obtain suitable surface roughness at a bonding surface.

[0062] The bonding layer **40** may be formed by an arbitrary appropriate method. For example, the bonding layer **40** may be formed by physical deposition such as sputtering, vacuum deposition, or ion beam assisted deposition (IAD), chemical deposition, or an atomic layer deposition (ALD) method. The film formation of the bonding layer **40** can be performed at a room temperature (25° C.) to 300° C., for example.

### A-4. Electrode Layers

[0063] Examples of materials used to constitute the electrode layers **30** to **33** include metal such as Pt, Au, Ti, Cr, Ni, Mo, Al, Ru, and SRO, and a compound and an oxide of these materials. One of these may be used alone, or two or more kinds thereof may be used in combination.

[0064] In one embodiment, materials constituting the electrode layers **30** to **33** are substantially the same. Specifically, the electrode layers **30** to **33** have substantially the same composition. In the composite substrate **140**, for example, the electrode layer **31** is constituted by metal (for example, Ti), and the electrode layer **32** is constituted by metal (for example, Ti). Such a configuration can be employed by constituting the piezoelectric layers by non-oriented polycrystalline bodies. In a case where the piezoelectric layers are formed through film formation, for example, adjacent layers (electrodes) have a function as seed crystal layers of the piezoelectric layers and are constituted by a material that has a predetermined physical property (for example, a lattice constant). On the other



hand, options for a material constituting the adjacent layers (electrodes) increases by constituting the piezoelectric layers by non-oriented polycrystalline bodies, and it is possible to select a material from a viewpoint of manufacturing efficiency and properties of the obtained composite substrate (piezoelectric element), for example.

[0065] The thickness of each of the electrode layers **30** to **33** that may function as layers in close contact with the adjacent layers is, for example, 1 nm or more and 100 nm or less, is preferably 3 nm or more and 50 nm or less, and is further preferably 5 nm or more and 20 nm or less.

[0066] The electrode layers **30** to **33** may be formed by an arbitrary appropriate method. For example, the electrode layers **30** to **33** may be formed by physical deposition such as sputtering, vacuum deposition, or ion beam assisted deposition (IAD). In one embodiment, the electrode layers **30** to **33** may be formed by performing sputtering using the same target (for example, a Ti target) under the same conditions. The film formation of the electrode layers **30** to **33** can be performed at a room temperature (25° C.) to 300° C., for example.

#### A-5. Manufacturing Method

[0067] The composite substrates **100** to **150** can be obtained by bonding (directly bonding) the piezoelectric layers constituted by sintered bodies, for example, or bonding the piezoelectric layers and the support substrate **50**.

[0068] FIG. **11** is a diagram illustrating an example of a manufacturing process of the composite substrate **110** according to the second embodiment, as an example of manufacturing processes of the composite substrates **100** to **150**. FIG. **11A** illustrates a film formation process in the manufacturing process of the composite substrate **110**. In the film formation process, the electrode layer **30** is formed through film formation on a surface of a piezoelectric substrate **70** that is a bulk sintered body after polarization processing.

[0069] FIG. **11B** illustrates a bonding process in the manufacturing process of the composite substrate **110**. In the bonding process, the electrode layer **30** formed on the piezoelectric substrate **70** in the film formation process in FIG. **11A** and the first piezoelectric layer **10** after polarization processing which also acts as a support substrate are brought into contact with each other after bonding surfaces thereof are activated by arbitrary appropriate activation processing, and are then pressurized at an ordinary temperature, thereby achieving direct bonding. At this time, disposition of the piezoelectric substrate **70** and the first piezoelectric layer **10** is determined such that the polarization direction of the piezoelectric substrate **70** and the polarization direction of the first piezoelectric layer **10** are mutually opposite directions.

[0070] In one embodiment, an element (for example, argon) that constitutes gas to be used for the activation processing is contained near a bonding interface between the electrode layer **30** and the first piezoelectric layer **10**. Specifically, an end portion of at least one of the electrode layer **30** and the first piezoelectric layer **10** facing each other via the bonding interface is an amorphous region (a region including an amorphous body; an amorphous layer) containing an element constituting gas to be used for the activation processing. The thickness of such an amorphous region is, for example, 2 nm to 30 nm. The concentration of argon in the amorphous region is, for example, 0.5 atm % to 30 atm %. Although a distribution state of argon in the amorphous region is not particularly limited, the concentration of argon increases toward the side of the bonding interface in the amorphous region, for example.

[0071] FIG. **11C** illustrates a polishing process in the manufacturing process of the composite substrate **110**. In the polishing process, the second piezoelectric layer **20** is formed by performing working such as grinding and polishing on the piezoelectric substrate **70** bonded to the first piezoelectric layer **10** via the electrode layer **30** in the bonding process in FIG. **11B** to a desired thickness. The composite substrate **110** is thus manufactured.

[0072] In one embodiment, working such as grinding and polishing is performed on the piezoelectric substrate **70** such that the obtained thickness of the second piezoelectric layer **20** exceeds 0.2  $\mu\text{m}$ . According to such an embodiment, it is possible to suppress occurrence of particle

shedding of crystals constituting the second piezoelectric layer **20** and peeling of the second piezoelectric layer **20** without weakening a coupling force of the grain boundary of the obtained second piezoelectric layer **20** and a coupling force with the support substrate due to a working load. [0073] Note that although FIG. **11** illustrates an example of the manufacturing process of the composite substrate **110** according to the second embodiment, the composite substrates described in the other embodiments can also be produced by a similar manufacturing process. However, since the first piezoelectric layer **10** and the second piezoelectric layer **20** are directly bonded in the composite substrate **100** according to the first embodiment, the film formation process in FIG. **11A** is omitted. Also, the bonding layer **40** is formed through film formation instead of the electrode layer **30**, the bonding layer **40** and the first piezoelectric layer **10** are directly bonded, and the amorphous layer is formed on the bonding interface therebetween in the film formation process in FIG. **11A** for the composite substrate **120** according to the third embodiment.

[0074] Furthermore, the first piezoelectric layer **10** with a desired thickness is formed on the support substrate **50**, and the amorphous layer is formed on the bonding interface between the support substrate **50** and the first piezoelectric layer **10** by performing the bonding process in FIG. **11B** and the polishing process in FIG. **11C** using the support substrate **50** and the piezoelectric substrate **70** for the composite substrate **130** according to the fourth embodiment. Thereafter, the second piezoelectric layer **20** with a desired thickness is formed on the first piezoelectric layer **10**, and the amorphous layer is formed on the bonding interface between the first piezoelectric layer **10** and the second piezoelectric layer **20** by performing the bonding process in FIG. **11B** and the polishing process in FIG. **11C** again. At this time, in order for the polarization direction of the piezoelectric substrate **70** to form the second piezoelectric layer **20** and the polarization direction of the first piezoelectric layer **10** to become mutually opposite directions as described above, the disposition thereof is determined. However, since the support substrate **50** and the first piezoelectric layer **10** are directly bonded, and the first piezoelectric layer and the second piezoelectric layer **20** are directly bonded in the composite substrate **130** according to the fourth embodiment, the film formation process in FIG. **11A** is omitted.

[0075] Also, the first piezoelectric layer **10** or the piezoelectric layer **11** with a desired thickness is formed on the support substrate **50** with the electrode layer **31** interposed therebetween, and the amorphous layer is formed on the bonding interface between the electrode layer **31** and the first piezoelectric layer **10** or on the bonding interface between the electrode layer **31** and the first piezoelectric layer **11** by performing the film formation process in FIG. **11A**, the bonding process in FIG. **11B**, and the polishing process in FIG. **11C** using the support substrate **50** and the piezoelectric substrate **70** for the composite substrate **140** according to the fifth embodiment and the composite substrate **150** according to the sixth embodiment. Thereafter, the second piezoelectric layer **20** or the piezoelectric layer **12** with a desired thickness is formed on the first piezoelectric layer **10** or the piezoelectric layer **11** with the electrode layer **32** interposed therebetween, and the amorphous layer is formed on the bonding interface between the electrode layer **32** and the second piezoelectric layer **20** or on the bonding interface between the electrode layer **32** and the second piezoelectric layer **12** by performing the film formation process in FIG. **11A**, the bonding process in FIG. **11B**, and the polishing process in FIG. **11C** again. At this time, in order for the polarization direction of the piezoelectric substrate **70** to form the second piezoelectric layer **20** and the polarization direction of the first piezoelectric layer **10** to become mutually opposite directions in the composite substrate **140** as described above, the disposition thereof is determined. On the other hand, the electrode layer **33** is further formed by performing the film formation process in FIG. **11A** after the piezoelectric layer **12** is formed in the composite substrate **150**.

[0076] In the composite substrates **100** to **150**, the surface of each layer (specifically, the support substrate **50**, the first piezoelectric layer **10**, the second piezoelectric layer **20**, the electrode layers **30** to **32**, or the bonding layer **40**) is a flat surface. Specifically, arithmetic mean roughness  $R_a$  of

the surface of each layer is, for example, 5 nm or less, is preferably 2 nm or less, is more preferably 1 nm or less, and is further preferably 0.3 nm or less. Examples of a method of flattening the surface of each layer include chemical mechanical polishing (CMP) and mirror polishing by lap polishing.

[0077] At the time of the film formation process and the bonding process, it is preferable to clean the surface of each layer to remove a residue of an abrasive, for example. Examples of a cleaning method include wet cleaning, dry cleaning, and scrub cleaning. Among these, scrub cleaning is preferable because it is possible to simply and efficiently perform cleaning. Specific examples of the scrub cleaning include a method of cleaning by a scrub cleaning machine using a solvent (for example, a mixed solution of acetone and isopropyl alcohol (IPA)) after using a cleaning agent (for example, the Sunwash series manufactured by Lion Corporation).

[0078] The activation processing is representatively performed by irradiation with a neutral beam. Preferably, a neutral beam is generated using an apparatus such as an apparatus described in Japanese Patent Laid-Open No. 2014-086400, and irradiation with the beam is performed, thereby performing the activation processing. Specifically, a fast atom beam source of a saddle field type is used as a beam source, inert gas such as argon or nitrogen is introduced into a chamber, and a high voltage is applied from a DC power source to an electrode. In this manner, electrons move due to an electric field of the saddle field type generated between the electrode (positive pole) and a casing (negative pole), and beams of atoms and ions by the inert gas are generated. Since the ion beam is neutralized by a grid among the beams that have reached the grid, a beam of neutral atoms is emitted from the fast atom beam source. The voltage at the time of the activation processing through the beam irradiation is preferably 0.5 kV to 2.0 kV, and the current at the time of the activation processing through the beam irradiation is preferably 50 mA to 200 mA.

[0079] The contact and the pressurization of the bonding surfaces are preferably performed in a vacuum atmosphere. The temperature at this time is representatively an ordinary temperature. Specifically, the temperature is preferably 20° C. or higher and 40° C. or lower, and is more preferably 25° C. or higher and 30° C. or lower. The pressure to be applied is preferably 100 N to 20000 N.

[0080] Although the piezoelectric substrate **70** provided with the electrode layer **30** through film formation and the first piezoelectric layer **10** are bonded to obtain the composite substrate **110** in the example in FIG. **11**, the present invention is not limited to such a mode. For example, the electrode layer **30** formed on the first piezoelectric layer **10** may be bonded to the piezoelectric substrate **70** after the electrode layer **30** is provided on the side of the first piezoelectric layer **10**. The same applies to the other embodiments.

[0081] In the bonding process, at least one of the first piezoelectric layer **10** and the second piezoelectric layer **20** is bonded to another layer, and the amorphous layer may thus be formed on the bonding interface between the piezoelectric layer and another layer. Specifically, in the composite substrate **100** according to the first embodiment and the composite substrate **130** according to the fourth embodiment, the amorphous layers may be formed on the bonding interfaces between the first piezoelectric layers **10** and the second piezoelectric layers **20**. Also, in the composite substrate **110** according to the second embodiment and the composite substrate **140** according to the fifth embodiment, the electrode layers **30** and **32** are disposed between the first piezoelectric layers **10** and the second piezoelectric layers **20**, and the amorphous layers may be formed on the bonding interfaces between the first piezoelectric layers **10** or the second piezoelectric layers **20** and the electrode layers **30** and **32**. Similarly, in the composite substrate **120** according to the third embodiment, the bonding layer **40** is disposed between the first piezoelectric layer **10** and the second piezoelectric layer **20**, and the amorphous layer may be formed on the bonding interface between the first piezoelectric layer **10** or the second piezoelectric layer **20** and the bonding layer **40**.

EXAMPLE

[0082] Hereinafter, an example of a composite substrate with a piezoelectric layer directly bonded to a support substrate will be specifically described as an example for verifying the structure of the composite substrate according to the present invention. Note that the following procedure was performed at a room temperature unless particularly stated otherwise.

[0083] PbZrO<sub>3</sub> powder, PbTiO<sub>3</sub> powder, Nb<sub>2</sub>O<sub>5</sub> powder, and ZnO powder were stirred and mixed by a ball mill using water as a dispersant, and the obtained mixture was dried and then calcined in the atmosphere (at 900° C. for 2 hours). Thereafter, wet milling was performed again by the ball mill for 20 hours, thereby obtaining powder with particle diameters of about 1 μm. Then, the powder was press-molded, thereby obtaining a molded article.

[0084] The obtained molded article was subjected to pre-sintering in the atmosphere at 1250° C. for 2 hours. After the sintering, the molded article was cooled in the atmosphere, thereby obtaining a pre-sintered body. The obtained pre-sintered body was buried in a container filled with mixed powder of PbO and ZrO<sub>2</sub>, the container with a lid thereon was placed in an internal-heating high-temperature high-pressure furnace, the temperature was raised from a room temperature to 1100° C. for 4.5 hours, and hot isostatic press processing (HIP method) was performed thereon.

Specifically, pressurization was performed up to 280 bar at 1000° C. and the pressure was raised from 280 bar to 600 bar for 1 hour after the point at which the temperature exceeded 1000° C. at the time of the raising of the temperature, the pre-sintered body was held at 1100° C. and 600 bar for 1 hour, and hot isostatic press processing was performed. A plate-shaped sintered body was thus obtained.

[0085] Electrodes were formed on the upper surface and the lower surface of the obtained sintered body, and polarization processing was performed by applying a predetermined voltage. Thereafter, beveling, grinding, and lap polishing were performed on the sintered body, thereby obtaining a wafer (piezoelectric substrate) that had a first surface and a second surface facing each other and had a diameter of 4 inches and a thickness of 500 μm.

[0086] The first surface of the obtained piezoelectric substrate was finished through chemical mechanical polishing (CMP) and was mirror-finished such that arithmetic mean roughness Ra became less than 2 nm. Here, the arithmetic mean roughness Ra was a value measured by an atomic force microscope (AFM) with a field of view of 10 μm×10 μm.

[0087] A Ti film with a thickness of 10 nm, a Pt film with a thickness of 100 nm, a Ti film with a thickness of 10 nm, and a silicon film with a thickness of 150 nm were formed in this order on the mirror-finished first surface of the piezoelectric substrate through sputtering. Thereafter, chemical mechanical polishing (CMP) was performed on the surface of the silicon film to obtain the arithmetic mean roughness Ra of 0.2 nm.

[0088] A silicon substrate (support substrate) that had an orientation flat portion and had a diameter of 4 inches and a thickness of 500 μm was prepared. Chemical mechanical polishing (CMP) was performed on the surface of the silicon substrate in advance, and the arithmetic mean roughness Ra was 0.2 nm.

[0089] Then, the piezoelectric substrate and the support substrate was directly bonded. Specifically, the surface (on the side of the silicon film) of the piezoelectric substrate and the surface of the support substrate were cleaned, both the substrates were then put into a vacuum chamber, the vacuum chamber was evacuated up to a 10<sup>-6</sup> Pa level, and the surfaces of both the substrates were then irradiated with a fast atom beam (acceleration voltage of 1 kV, Ar flow rate of 27 sccm) for 120 seconds. After the irradiation, the surfaces of both the substrates irradiated with the beam were overlaid, and both the substrates were pressurized at 10000 N for 2 minutes and were bonded, thereby obtaining a bonded body.

[0090] Then, the second surface of the piezoelectric substrate in the obtained bonded body was ground and polished, thereby obtaining a composite substrate including a piezoelectric layer with a thickness of 10 μm.

<Tem Observation>

[0091] Transmission electron microscope (TEM) observation (50,000 folds, 400,000 folds, and 2,000,000 folds) of the section of the composite substrate produced as described above was performed. Observation photographs are shown in FIGS. 12A, 12B, and 12C. Note that observation samples were produced from the obtained composite substrate by the FIB method for the sectional TEM observation.

<Edx Analysis>

[0092] In EDX analysis of the section of the composite substrate, the concentration of argon in the layer (the amorphous layer formed through the activation processing) indicated by the arrow in FIG. 12C was 3.0 atm %.

[0093] In the example, it was possible to confirm the structure of the composite substrate in which the piezoelectric layer with the thickness of 10  $\mu\text{m}$  and the silicon substrate that was the support substrate were directly bonded with the metal films (the Ti films and the Pt film) that acted as the electrode layers and the silicon film that acted as the bonding layer interposed therebetween. In the composite substrate, an amorphous layer by the activation processing was formed on the bonding interface (the part indicated by the arrow in FIG. 12C) between the support substrate and the bonding layer. Therefore, it was possible to confirm that the composite substrates **100** to **150** described above were able to be produced through the similar processes.

[0094] According to the embodiments of the present invention described above, the following effects and advantages are achieved.

[0095] (1) Each of the composite substrates **100** to **150** includes the first piezoelectric layer **10** and the second piezoelectric layer **20** that is disposed to be stacked on the first piezoelectric layer, and the amorphous layer is formed on the bonding interface between at least one of the first piezoelectric layer **10** and the second piezoelectric layer **20** and another layer. In other words, at least one of the first piezoelectric layer **10** and the second piezoelectric layer **20** is directly bonded to another layer. It is thus possible to provide a composite substrate that has the piezoelectric layers capable of achieving both size reduction and a piezoelectric property.

[0096] (2) In each of the composite substrates **100** and **130**, the amorphous layer is formed on the bonding interface between the first piezoelectric layer **10** and the second piezoelectric layer **20**. In other words, the first piezoelectric layer **10** and the second piezoelectric layer **20** are directly bonded to each other. Also, each of the composite substrates **110**, **120**, **140**, and **150** includes the electrode layer **30** or **32** or the bonding layer **40** that is disposed between the first piezoelectric layer **10** (piezoelectric layer **11**) and the second piezoelectric layer **20** (piezoelectric layer **12**), and the amorphous layer is formed on the bonding interface between the first piezoelectric layer **10** (piezoelectric layer **11**) or the second piezoelectric layer **20** (piezoelectric layer **12**) and the electrode layer **30** or **32** or the bonding layer **40**. In other words, the first piezoelectric layer **10** (piezoelectric layer **11**) or the second piezoelectric layer **20** (piezoelectric layer **12**) and the electrode layer **30** or **32** or the bonding layer **40** are directly bonded to each other. Furthermore, each of the composite substrates **130**, **140**, and **150** includes the support substrate **50** that supports the first piezoelectric layer **10** (piezoelectric layer **11**) and the second piezoelectric layer **20** (piezoelectric layer **12**). It is thus possible to produce the composite substrates **100** to **150** with arbitrary layer structures.

[0097] (3) The support substrate **50** may be constituted by any of silicon, SOI, sialon, sapphire, cordierite, mullite, glass, quartz, crystal, alumina, stainless steel, an iron-nickel alloy (Alloy 42), and brass. It is thus possible to constitute the support substrate **50** by an arbitrary material in accordance with purposes.

[0098] (4) The thickness of at least one of the first piezoelectric layer **10** and the second piezoelectric layer **20** is preferably 50  $\mu\text{m}$  or less. It is thus possible to achieve size reduction when piezoelectric actuators are constituted using the composite substrates **100** to **150**.

[0099] (5) The polarization directions of the first piezoelectric layer **10** and the second piezoelectric layer **20** are mutually opposite directions. It is thus possible to form a composite substrate with a

bimorph structure.

[0100] (6) Each of the first piezoelectric layer **10** and the second piezoelectric layer **20** may be constituted by any of PZT, PMN-PT, barium titanate, lead titanate, lead metaniobate, bismuth titanate, KNN, KNN-LN, BT-BNT-BKT, LiTaO<sub>3</sub>, LiNbO<sub>3</sub>, and crystal. Also, each of the first piezoelectric layer **10** and the second piezoelectric layer **20** may be constituted by a polycrystalline body. It is thus possible to constitute the first piezoelectric layer **10** and the second piezoelectric layer **20** by an arbitrary material in accordance with purposes.

[0101] Note that the present invention is not limited to the above embodiments and can be implemented using arbitrary components without departing from the gist thereof.

[0102] The aforementioned embodiments and modifications are only examples, and the present invention is not limited to the content thereof unless features of the present invention are lost. Also, although various embodiments and modifications have been described above, the present invention is not limited to the content thereof. Other aspects that are conceivable within the scope of the technical idea of the present invention are also included in the scope of the present invention.

## Claims

1. A composite substrate comprising: a first piezoelectric layer; a second piezoelectric layer that is disposed to be stacked on the first piezoelectric layer; and a support substrate that supports the first piezoelectric layer and the second piezoelectric layer, wherein an amorphous layer is formed on a bonding interface between at least one of the first piezoelectric layer and the second piezoelectric layer and another layer, the second piezoelectric layer, the first piezoelectric layer, and the support substrate are stacked in this order, and each of the first piezoelectric layer and the second piezoelectric layer is constituted by a monocrystal body or a non-oriented polycrystalline body.
2. A composite substrate comprising: a first piezoelectric layer; and a second piezoelectric layer that is disposed to be stacked on the first piezoelectric layer, wherein an amorphous layer is formed on a bonding interface between at least one of the first piezoelectric layer and the second piezoelectric layer and another layer, polarization directions of the first piezoelectric layer and the second piezoelectric layer are mutually opposite directions, and each of the first piezoelectric layer and the second piezoelectric layer is constituted by a monocrystal body or a non-oriented polycrystalline body.
3. The composite substrate according to claim 1, wherein the amorphous layer is formed on a bonding interface between the first piezoelectric layer and the second piezoelectric layer.
4. The composite substrate according to claim 1, comprising: an electrode layer or a bonding layer that is disposed between the first piezoelectric layer and the second piezoelectric layer, wherein the amorphous layer is formed on a bonding interface between the first piezoelectric layer or the second piezoelectric layer and the electrode layer or the bonding layer.
5. The composite substrate according to claim 1, wherein the support substrate is constituted by any of silicon, SOI, sialon, sapphire, cordierite, mullite, glass, quartz, crystal, alumina, stainless steel, an iron-nickel alloy (Alloy 42), and brass.
6. The composite substrate according to claim 1, wherein at least one of the first piezoelectric layer and the second piezoelectric layer has a thickness of 50  $\mu\text{m}$  or less.
7. The composite substrate according to claim 1, wherein polarization directions of the first piezoelectric layer and the second piezoelectric layer are mutually opposite directions.
8. The composite substrate according to claim 1, wherein each of the first piezoelectric layer and the second piezoelectric layer is constituted by a non-oriented polycrystalline body of any of PZT, PMN-PT, barium titanate, lead titanate, lead metaniobate, bismuth titanate, KNN, KNN-LN, and BT-BNT-BKT, or is constituted by a monocrystal body of any of LiTaO<sub>3</sub>, LiNbO<sub>3</sub>, and crystal.
9. A composite substrate comprising: a first piezoelectric layer; and a second piezoelectric layer that is disposed to be stacked on the first piezoelectric layer, wherein at least one of the first

piezoelectric layer and the second piezoelectric layer is directly bonded to another layer, and each of the first piezoelectric layer and the second piezoelectric layer is constituted by a monocrystal body or a non-oriented polycrystalline body.

**10.** The composite substrate according to claim 9, wherein the first piezoelectric layer and the second piezoelectric layer are directly bonded to each other.

**11.** The composite substrate according to claim 9, comprising: an electrode layer or a bonding layer that is disposed between the first piezoelectric layer and the second piezoelectric layer, wherein the first piezoelectric layer or the second piezoelectric layer and the electrode layer or the bonding layer are directly bonded to each other.

**12.** The composite substrate according to claim 1, wherein at least one of the first piezoelectric layer and the second piezoelectric layer is constituted by a non-oriented polycrystalline body.

**13.** The composite substrate according to claim 12, wherein the non-oriented polycrystalline body is a sintered body.

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