

TARGET MEMBER FOR PHYSICAL VAPOR DEPOSITION, SPUTTERING TARGET
MEMBER, AND METHODS FOR PRODUCING PHYSICALLY VAPOR-DEPOSITED FILM
AND LAYERED STRUCTURE

5

TECHNICAL FIELD

The present invention relates to a target member for physical vapor deposition, and a sputtering target member, and methods for producing a physically vapor-deposited film and a layered structure.

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BACKGROUND ART

In recent years, a magnetic tunnel junction (MTJ) element has attracted attention as a magnetic recording element that improves the recording density of a magnetic recording apparatus. An MTJ element has a structure in which a tunnel barrier layer is sandwiched by two ferromagnetic layers, namely it has a three-layered structure of ferromagnetic layer/tunnel barrier layer/ferromagnetic layer. As a conventional tunnel barrier layer, an Al oxide film (amorphous AlO_x film) having an amorphous structure or a (001) oriented crystalline MgO film is used. However, the amorphous AlO_x film is not suitable for the tunnel barrier layer of an MTJ element, because it has a high junctional resistance with a ferromagnetic layer, a large interface roughness with the ferromagnetic layer to give large dispersion of characteristics, and a small tunnel magnetoresistance ratio (TMR ratio). On the other hand, the crystalline MgO film is suitable for the tunnel barrier layer of an MTJ element, because it has a smaller tunnel resistance (TR) with respect to a ferromagnetic substance having a bcc crystal structure such as Fe or FeCo, and a large TMR ratio. Therefore, it can be expected that the performance of an MTJ element is improved so as to downsize the MTJ element and to enhance further the recording density of a magnetic recording apparatus provided with the MTJ element. However, since MgO is easily hydrated, it may react with moisture in the atmosphere, or the like to form a hydroxide on the surface, and therefore there is a concern that the crystalline MgO film or a MgO sputtering target member for forming the same may be deteriorated.

Patent Document 1 describes a spinel structure MgAl_2O_4 film in which Al is added to a crystalline MgO film. The spinel structure MgAl_2O_4 film is very much expected as the tunnel barrier layer of an MTJ element, because it has been found that its TR is 10 times or more lower than that of an amorphous AlO_x film, a higher TMR ratio can be obtained, and the film is able to form an epitaxial tunnel junction with few defects because its lattice matching with respect to a ferromagnetic substance such as a Co-based full Heusler alloy or a CoFe alloy is better than that of a crystalline MgO film.

RELATED-ART DOCUMENT

10 PATENT DOCUMENT

Patent Document 1: JP-B-5586028

SUMMARY OF THE INVENTION

TECHNICAL PROBLEM

15 Meanwhile, the method for producing a spinel structure MgAl_2O_4 film described in Patent Document 1 is as follows. That is, a plasma oxidation treatment is performed after a Mg film and an Al film are layered one on another by sputtering on a ferromagnetic underlayer, or a plasma oxidation treatment is performed after a MgAl_2 alloy film is formed on a ferromagnetic underlayer by sputtering a MgAl_2 alloy, so that the metal film or the alloy film is oxidized and
20 crystallized to form a spinel structure MgAl_2O_4 film.

With respect to the method for producing a spinel structure MgAl_2O_4 film described in Patent Document 1, since a plasma oxidation treatment is performed after forming a metal film or an alloy film, degradation due to oxidation of the ferromagnetic underlayer is feared. Also, generation of defects is feared because as a result of oxidation and crystallization of the metal
25 film or the alloy film, penetration of oxygen atoms into the metal film or the alloy film, and rearrangement of metal atoms in the metal film or the alloy film are necessary. For this reason, removal of defects from the spinel structure MgAl_2O_4 film, or an epitaxial tunnel junction may not be sufficient, and it is feared that there is a limit in downsizing of an MTJ element, or densification of a magnetic recording apparatus.

Objects of first and second aspects of the present invention are to provide a target member for physical vapor deposition and a sputtering target member, in which deterioration of a underlayer due to oxidation is little in forming a physically vapor-deposited film, there appear few defects at a junction between the physically vapor-deposited film and the underlayer, the
5 lattice matching between the physically vapor-deposited film and the underlayer is good, and further deterioration in quality due to hydration is little due to a low hydration tendency of the target members and the physically vapor-deposited film. An object of a third aspect of the invention is to provide a physically vapor-deposited film, in which deterioration of a underlayer due to oxidation is little, there appear few defects at a junction with the underlayer, lattice
10 matching with the underlayer is good, and further deterioration in quality due to hydration is little. An object of a fourth aspect of the invention is to provide a layered structure, for which downsizing can be expected through improvement in the TMR ratio because deterioration of a underlayer due to oxidation is little, there appear few defects at a junction between a physically vapor-deposited film and the underlayer, the lattice matching between the physically vapor-
15 deposited film and the underlayer is good, and further deterioration in quality due to hydration is little.

SOLUTION TO PROBLEM

(1) According to a first aspect of the invention, there is provided a target member for
20 physical vapor deposition comprising Mg, M (M is a trivalent metal element), and O as main components, wherein

a molar ratio of the Mg to the M in terms of the oxides of MgO and M_2O_3 respectively is 70:30 to 10:90.

A physically vapor-deposited film that is physically deposited on a underlayer using a
25 target member for physical vapor deposition containing Mg, M (M is a trivalent metal element), and O as main components does not require an oxidation treatment after the film formation, because necessary oxygen (O) is supplied from the target member for physical vapor deposition. In consequence, deterioration of the underlayer due to oxidation is little. Further, since oxidation or crystallization of a metal film or an alloy film is unnecessary, it is possible to reduce

defects to be generated in a junction between the physically vapor-deposited film and the underlayer. In addition, a physically vapor-deposited film exhibiting good lattice matching with the underlayer may be formed by physical vapor deposition on the underlayer using a target member for physical vapor deposition in which the molar ratio of Mg to M in terms of the oxides of MgO and M₂O₃ respectively is 70:30 to 10:90. Furthermore, since the target member for physical vapor deposition and the physically vapor-deposited film are not MgO, deterioration in quality due to hydration is little, and the stability such as hydration resistance can be excellent.

(2) According to a second aspect of the invention, there is provided a target member for physical vapor deposition comprising Mg, M (M is a trivalent metal element), and O as main components, and comprising a crystal phase having a spinel structure.

A physically vapor-deposited film physically vapor deposited on a underlayer using a target member for physical vapor deposition containing Mg, M (M is a trivalent metal element), and O as main components does not require an oxidation treatment after the film formation, because necessary oxygen (O) is supplied from the target member for physical vapor deposition. In consequence, deterioration of the underlayer due to oxidation is little. Further, since oxidation or crystallization of a metal film or an alloy film is unnecessary, it is possible to reduce defects to be generated in a junction between the physically vapor-deposited film and the underlayer. Furthermore, a physically vapor-deposited film exhibiting good lattice matching with the underlayer may be formed by physical vapor deposition on the underlayer using a target member for physical vapor deposition including a crystal phase having a spinel structure. Since the target member for physical vapor deposition and the physically vapor-deposited film are not MgO, deterioration in quality due to hydration is little, and the stability such as hydration resistance can be excellent.

(3) In the first or the second aspect of the invention, M is preferably one or two selected from the group consisting of Al and Ga. A physically vapor-deposited film having better lattice matching with the underlayer may be formed by physical vapor deposition on a underlayer using a target member for physical vapor deposition, in which M is one or two selected from the group consisting of Al and Ga.

(4) In the first or the second aspect of the invention, the light transmittance for a thickness of 2 mm is preferably 60% or less. As a use of the composition containing Mg, M (especially, Al), and O as main components, a window material for electrical equipment or a production vessel is well known. However the target member for physical vapor deposition does not
5 require a light transmittance as high as that of the window material. Therefore, by configuring a target member for physical vapor deposition allowing a light transmittance for a thickness of 2 mm of 60% or less, a target member for physical vapor deposition can be manufactured more simply and inexpensively.

(5) In the first or the second aspect of the invention, the dielectric loss at 10 GHz is
10 preferably 45000 GHz or more in terms of f·Q value. The dielectric loss of the target member for physical vapor deposition at 10 GHz in terms of f·Q value of 45000 GHz or more reflects the fact that there are few defects and unavoidable impurities in the target member for physical vapor deposition. When physical vapor deposition is performed using such a target member for physical vapor deposition, a uniform physically vapor-deposited film in which there exist fewer
15 defects and unavoidable impurities can be formed.

(6) In the first or the second aspect of the invention, Whiteness is preferably 30 or more. Whiteness of 30 or more reflects the fact that there exist few unavoidable impurities and defects in the target member for physical vapor deposition. When physical vapor deposition is performed using such a target member for physical vapor deposition, a uniform physically
20 vapor-deposited film in which there exist fewer unavoidable impurities and defects can be formed.

(7) In the first or the second aspect of the invention, the target member for physical vapor deposition is suitable for a sputtering target member.

(8) The third aspect of the invention relates to a method for producing a physically vapor-deposited film, comprising physically vapor depositing the physically vapor-deposited film on a
25 underlayer using the target member for physical vapor deposition according to the first or the second aspect.

A physically vapor-deposited film physically vapor-deposited on a underlayer using the target member for physical vapor deposition according to the first or the second aspect does not

require an oxidation treatment after the film formation, because necessary oxygen (O) is supplied from the target member for physical vapor deposition. In consequence, deterioration of the underlayer due to oxidation is little. Further, since oxidation or crystallization of a metal film or an alloy film is unnecessary, it is possible to reduce defects to be generated in a junction
5 between the physically vapor-deposited film and the underlayer. Furthermore, a physically vapor-deposited film exhibiting good lattice matching with the underlayer may be formed by physical vapor deposition on the underlayer using a target member for physical vapor deposition in which the molar ratio of the Mg to the M in terms of the oxides of MgO and M_2O_3 respectively is 70:30 to 10:90, or using a target member for physical vapor deposition including
10 a crystal phase having a spinel structure. Since the physically vapor-deposited film is not MgO, deterioration in quality due to hydration is little, and the stability such as hydration resistance is excellent.

(9) The fourth aspect of the invention relates to a method for producing a layered structure, comprising: physically vapor depositing a physically vapor-deposited film on a
15 underlayer using the target member for physical vapor deposition according to the first or the second aspect of the invention, and forming a ferromagnetic layer on the physically vapor-deposited film, wherein the underlayer is a ferromagnetic layer, and the physically vapor-deposited film is a tunnel barrier layer.

A physically vapor-deposited film physically vapor-deposited on a underlayer using the
20 target member for physical vapor deposition according to the first or the second aspect does not require an oxidation treatment after the film formation, because necessary oxygen (O) is supplied from the target member for physical vapor deposition. In consequence, the underlayer deteriorates little due to oxidation. Further, since oxidation or crystallization of the metal film or the alloy film is unnecessary, it is possible to reduce defects to be generated in the junction
25 between the physically vapor-deposited film and the underlayer. Furthermore, a physically vapor-deposited film exhibiting good lattice matching with the underlayer may be formed by physical vapor deposition using a target member for physical vapor deposition in which the molar ratio of the Mg to the M in terms of the oxides of MgO and M_2O_3 respectively is 70:30 to 10:90, or using a target member for physical vapor deposition including a crystal phase having a

spinel structure. When the underlayer is a ferromagnetic layer and the physically vapor-deposited film is a tunnel barrier layer, a layered structure is formed in which the tunnel barrier layer is sandwiched between two ferromagnetic layers. Since the layered structure improves the TMR ratio, it is possible to produce a magnetic tunnel junction element downsized from the conventional one. Further, since the physically vapor-deposited film is not MgO, deterioration in quality due to hydration is little, and the stability such as hydration resistance is excellent.

DESCRIPTION OF EMBODIMENTS

The target member for physical vapor deposition of the invention can be used for a known physical vapor deposition method, such as a resistance heating evaporation method, a sputtering method, an electron-beam evaporation method, a molecular beam epitaxy method, an ion plating deposition method, and a laser ablation method. As an embodiment of the invention, a sputtering target member used for a sputtering method will be described below. The embodiment described below does not limit the contents of the invention described in the claims without due cause, and not all the components described in the embodiment are indispensable for the solution to the problem to be solved by the invention.

(1) Sputtering target member

A sputtering target member of the present embodiment contains Mg, M (M is a trivalent metal element), and O as main components. The sputtering target member may contain a subcomponent in addition to the main components. A sputtered film formed by sputtering a sputtering target member containing Mg, M (M is a trivalent metal element), and O as main components does not require an oxidation treatment after the film formation, because necessary oxygen (O) is supplied from the sputtering target member. In consequence, deterioration of the underlayer due to oxidation is little. Further, since oxidation or crystallization of a metal film or an alloy film is unnecessary, it is possible to reduce defects to be generated in a junction between the sputtered film and the underlayer. Since the sputtering target member and the sputtered film are not MgO, deterioration in quality due to hydration is little, and the stability such as hydration resistance is excellent.

The molar ratio of the Mg to the M contained in the sputtering target member in terms of the oxides of MgO and M_2O_3 respectively is preferably 70:30 to 10:90, more preferably 65:35 to 20:80, further preferably 60:40 to 30:70, and especially preferably 55:45 to 40:60. By adjusting the molar ratio of the Mg and the M contained in the sputtering target member in terms of the oxides of MgO and M_2O_3 respectively, the crystal phase or the lattice constants of the sputtered film can be controlled. Therefore, a sputtered film exhibiting good lattice matching with the underlayer may be formed.

The sputtering target member preferably includes a crystal phase having a spinel structure. By sputtering a sputtering target member including a crystal phase having a spinel structure, a sputtered film exhibiting good lattice matching with the underlayer may be formed.

M contained in the sputtering target member is preferably one or two selected from the group consisting of Al and Ga. By sputtering the sputtering target member in which M is one or two selected from the group consisting of Al and Ga, a sputtered film exhibiting better lattice matching with the underlayer may be formed.

The light transmittance of the sputtering target member for a thickness of 2 mm is preferably 60% or less, and more preferably 45% or less. As a use of a composition containing Mg, M (especially, Al), and O as main components, a window material for electrical equipment or a production vessel is well known. However the sputtering target member does not require a light transmittance as high as that of the window material. Therefore, by regulating the light transmittance into the above range, a sputtering target member can be manufactured more simply and inexpensively.

The content of unavoidable impurities in the sputtering target member is at most 0.5 mass%, preferably 0.1 mass% or less, and more preferably 0.01 mass% or less. By sputtering a sputtering target member containing little unavoidable impurities, a more uniform sputtered film containing less unavoidable impurities and defects can be formed.

The dielectric loss of a sputtering target member at 10 GHz in terms of f·Q value is preferably 45000 GHz or more, more preferably 50000 GHz or more, further preferably 70000 GHz or more, and especially preferably 80000 GHz or more. A large f·Q value of dielectric loss reflects there being few defects and unavoidable impurities in the sputtering target member.

By sputtering such a sputtering target member, a uniform sputtered film in which there exist fewer defects and unavoidable impurities can be formed. The dielectric loss is expressed by $\tan \delta$ or its inverse, namely a quality factor Q value ($= 1/\tan \delta$), and varies with the frequency. Therefore, an f·Q value that is the product of frequency f and Q value is also used. The larger the f·Q value is, the smaller the dielectric loss is.

Whiteness of the sputtering target member is preferably 30 or more, more preferably 50 or more, further preferably 60 or more, and especially preferably 65 or more. High Whiteness reflects there existing few unavoidable impurities and defects in the sputtering target member. By sputtering such a sputtering target member, a uniform sputtered film in which there exist fewer defects and unavoidable impurities can be formed. As Whiteness, L^* in the system of CIE 1976 (L^* , a^* , b^*) is used. Whiteness closer to 100 means closer to pure white.

The surface roughness of the sputtering target member is preferably 5 μm or less, more preferably 2 μm or less, and further preferably 1 μm or less. As the surface roughness of a sputtering target member becomes smaller, in other words, as the surface becomes smoother, the surface of the sputtering target member can be sputtered more uniformly to form a uniform sputtered film containing less defects.

The thickness and diameter of the sputtering target member may be changed to a desired thickness and diameter corresponding to a sputtering apparatus, and as an example, the thickness may be 2.0 mm or less.

There is no particular restriction on the material of the sputtering target member as long as the material is uniform and dense, and it is preferably composed of a sintered body. Thereby a uniform and dense sputtering target member can be manufactured more simply and inexpensively than by other solid producing methods such as a melt-solidification method.

The relative density of the sintered body is preferably 95 mass% or more, more preferably 98 mass% or more, further preferably 99 mass% or more, and especially preferably 99.5 mass% or more. As the relative density of the sintered body becomes higher, the sputtering target member becomes more dense and uniform to form a uniform sputtered film containing less defects.

The three-point bending strength of the sintered body is preferably 230 MPa or more, more preferably 250 MPa or more, further preferably 300 MPa or more, and especially preferably 320 MPa or more. As the bending strength of the sintered body becomes higher, the sputtering target member becomes more dense and uniform to form a uniform sputtered film containing less defects. In this regard, JIS R1601 is used as a method for measuring the three-point bending strength.

There is no particular restriction on the average grain diameter of the crystal grains constituting the sintered body as long as the sintered body can be made uniform and dense, and the average grain diameter is preferably 1 to 100 μm , more preferably 2 to 80 μm , further preferably 2 to 60 μm , and especially preferably 2 to 50 μm , when a Heywood equivalent diameter that is obtained by converting the area of a grain into a circle is used as the grain diameter. By sputtering a uniform and dense sputtering target member, a uniform sputtered film containing less defects may be formed.

There is no particular restriction on the D90/D10 of the crystal grains constituting the sintered body as long as the sintered body can be made uniform and dense, and the D90/D10 is preferably 4 or less, more preferably 3 or less, further preferably 2.5 or less, and especially preferably 2.3 or less. By sputtering a uniform and dense sputtering target member, a uniform sputtered film containing less defects may be formed.

The grain diameters of 200 crystal grains are measured, and the value (D50) of the grain diameter distribution at 50% (based on number) is used as the average grain diameter. Similarly, as D10 and D90, the values of the grain diameter distribution at 10% and 90% (based on number) are used.

(2) Method for producing sputtering target member

A method for producing a sputtering target member of the present embodiment includes a raw material mixing step of weighing and mixing powders of raw materials to prepare a slurry; a drying and granulating step of drying and granulating the slurry to yield a granulated powder; a molding step of molding the granulated powder to yield a molded body; a sintering step of sintering the molded body to yield a sintered body; and an outline machining step of machining

the outline of the sintered body to yield a sputtering target member. The method for producing a sputtering target member will be described in more detail below.

(2-1) Raw material mixing step

As raw materials for a sputtering target member, powders of MgO and M_2O_3 (M is a trivalent metal element) can be used. The M is preferably one or two selected from the group consisting of Al and Ga. The raw material may contain MgO and M_2O_3 as main components, and may further contain a subcomponent according to need. It is preferable that the purity of the powders of the raw materials is as high as possible, and it is at least 99.5 mass% or more, preferably 99.9 mass% or more, more preferably 99.99 mass% or more, and further preferably 99.999 mass% or more. When a raw material powder with a high purity is used, a sputtering target member containing only a small amount of unavoidable impurities can be obtained. By sputtering this sputtering target member, a uniform sputtered film that contains less unavoidable impurities or defects may be formed.

As the average grain diameter (D50) of the raw material powder becomes smaller, sintering is promoted and a more uniform and dense sputtering target member is obtained, and therefore it is at least 100 μm or less, preferably 1 μm or less, and more preferably 0.5 μm or less in terms of a Heywood equivalent diameter that is obtained by converting the area of a grain into a circle.

The raw material for the sputtering target member is not limited to the oxides of MgO and M_2O_3 , and it is also possible to use a compound that may be converted into an oxide in a production process, such as a carbonate and a nitrate.

Powders of the raw materials are weighed. The molar ratio of the Mg to the M in terms of the oxides of MgO and M_2O_3 respectively is preferably 70:30 to 10:90, more preferably 65:35 to 20:80, further preferably 60:40 to 30:70, and especially preferably 55:45 to 40:60. By adjusting the molar ratio of the Mg to the M, it is possible to control the crystal phase and the lattice constants of a sputtered film. Therefore, a sputtered film exhibiting good lattice matching with the underlayer may be formed.

Powders of the raw materials are mixed to yield a slurry. There is no particular restriction on the mixing method as long as the powders of the raw materials can be uniformly

mixed, and, for example, wet ball milling is suitable. In wet ball milling, the powders of the raw materials, a dispersion medium, and balls are placed in a vessel, and mixed (wet mixing). As the dispersion medium, water, or an organic solvent such as alcohol and methanol, may be used. There is no particular restriction on the mass ratio of the raw materials to the dispersion medium as long as the powders of the raw materials can be uniformly mixed, and the mass ratio is generally often 15:85 to 75:25. For the sake of mixing uniformly the powders of the raw materials, a dispersing agent may be further added. There is no particular restriction on the dispersing agent as long as the dispersing agent is degraded and removed in a degreasing or sintering step described later. There is no particular restriction on the mixing time as long as the powders of the raw materials can be uniformly mixed, and the mixing time is preferably 10 hours or more. Unless the powders of the raw materials are uniformly mixed, unevenness in the composition or in the density is prone to appear in the sputtering target member to decrease the strength. Further, in a case where such a sputtering target member is sputtered, abnormal electrical discharge is prone to occur in sputtering, and a non-uniform sputtered film having many defects is likely to be formed.

The raw material mixing step may further include a drying step of drying the slurry after mixing the powders of the raw materials to yield a dry powder; a heat treatment step of heat-treating the dry powder to yield a heat-treated powder, in which part or all of the raw materials is composed of a composite oxide; and a milling step of placing the heat-treated powder, methanol as a dispersion medium, and alumina balls in a vessel, and wet-milling the content to obtain a slurry. Since the reaction of the powders of the raw materials advances by the heat treatment, the sputtering target member obtained in the subsequent sintering step is prone to include a crystal phase having a spinel structure. The grain diameter of the heat-treated powder is preferably as small as possible so as to promote sintering in the subsequent sintering step. For this reason, the milling step may further introduce dry-milling before the wet-milling.

(2-2) Drying and granulating step

The slurry prepared in the raw material mixing step is dried and granulated to yield a granulated powder suitable for molding. If necessary, a molding aid may be added to the slurry before drying. There is no particular restriction on the molding aid, and generally poly(vinyl

alcohol) (PVA), poly(ethylene glycol) (PEG), Selosol, paraffin, or the like is often used. There is no particular restriction on the drying method, and suitable examples thereof include a rotary evaporator and a spray dryer. The purity of the granulated powder is preferably as high as possible, and is at least 99.5 mass%, more preferably 99.9 mass%, and further preferably 99.99 mass% or more.

(2-3) Molding step

The granulated powder is molded to yield a molded body having a predetermined shape. As the molding method, known molding methods, such as uniaxial molding using a mold, and CIP (cold isostatic pressing) molding, may be used singly or in combination. There is no particular restriction on the molding pressure as long as a good molded body can be obtained, and the molding pressure is generally preferably 100 MPa or more. In this regard, in a case where HP (hot uniaxial pressing) sintering, or HIP (hot isostatic pressing) sintering is performed in the sintering step described later, the molding step may be omitted, or its molding pressure may be reduced.

(2-4) Sintering step

The molded body is sintered to yield a sintered body. By sintering, a uniform and dense sputtering target member can be produced more simply and inexpensively than by other solid producing methods. As the sintering method, known sintering methods, such as pressureless sintering, HP sintering, and HIP sintering, may be performed singly or in combination. There is no particular restriction on the sintering temperature as long as a sintered body can be obtained, and the sintering temperature is preferably 1800°C or less because the pressureless sintering can be performed in the atmosphere. By the HP sintering or HIP sintering, a high density sintered body may be obtained at a sintering temperature lower than in the case of pressureless sintering. In a case where a dispersing agent or a molding aid is contained in the granulated powder, it is preferable to carry out degreasing before sintering in order to degrade and remove these additives. There is no particular restriction on the degreasing temperature, and a temperature and a temperature increase rate at which the dispersing agent and the molding aid are completely degraded and removed are preferable. As for the method for producing a sputtering target

member, another solid production method such as melt solidification may be used as long as a uniform and dense sputtering target member can be produced.

(2-5) Outline machining step

The sintered body is processed into a desired shape to yield a sputtering target member.

- 5 Known methods such as cutting, grinding, and polishing may be used as the method of outline machining. The sputtering target member is bonded with a backing plate, and used for sputtering as a sputtering target.

(3) Application of sputtering target member

- 10 As an application example of the sputtering target member of this embodiment, a layered structure in which a tunnel barrier layer is sandwiched between two ferromagnetic layers, namely an MTJ element having a three-layered structure of under ferromagnetic layer/tunnel barrier layer/upper ferromagnetic layer, will be described.

- 15 An MTJ element is formed on a substrate. As the substrate, for example, a spinel MgAl_2O_4 single crystal, a Si single crystal, a GaAs single crystal, or thermally-oxidized Si may be used. On the surface of the substrate, if necessary, a buffer layer composed of, for example, MgO may be formed.

- 20 An under ferromagnetic layer (underlayer), a tunnel barrier layer, and an upper ferromagnetic layer are sequentially formed on the substrate. The under ferromagnetic layer, the tunnel barrier layer, and the upper ferromagnetic layer can be formed, for example, by sputtering a Co-based full Heusler alloy (for example, $\text{Co}_2\text{FeAl}_{0.5}\text{Si}_{0.5}$), the sputtering target member of this embodiment, and a CoFe alloy (for example, $\text{Co}_{75}\text{Fe}_{25}$ alloy) respectively. A tunnel barrier layer (sputtered film) formed by sputtering the sputtering target member of this embodiment does not require an oxidation treatment after the film formation, because necessary oxygen (O) is supplied from the sputtering target member. In consequence, deterioration of the
- 25 under ferromagnetic layer due to oxidation is little. Further, since oxidation or crystallization of the tunnel barrier layer is unnecessary, it is possible to reduce defects to be generated in a junction between the tunnel barrier layer and the under ferromagnetic layer. Furthermore, the tunnel barrier layer exhibits good lattice matching with the under ferromagnetic layer composed of the Co-based full Heusler alloy, and the upper ferromagnetic layer composed of the CoFe

alloy. In consequence, the TMR ratio of an MTJ element having the under ferromagnetic layer/tunnel barrier layer/upper ferromagnetic layer is improved, and therefore an MTJ element having a size smaller than the conventional one can be produced. A magnetic recording apparatus provided with the MTJ element can further increase the recording density.

5 Furthermore, since the tunnel barrier layer is not MgO, deterioration in quality due to hydration is little, and the stability such as hydration resistance is excellent.

As those skilled in the art will easily understand, the use of the sputtering target member of this embodiment is not limited to the sputtering method, and it can be used for a known physical vapor deposition method, such as a resistance heating evaporation method, an electron-
10 beam evaporation method, a molecular beam epitaxy method, an ion plating deposition method, and a laser ablation method.

EXAMPLES

Examples of the invention will be described in detail below.

15 (1) Production of sputtering target member

(1-1) Example 1

As raw materials, a powder of MgO having an average grain diameter of 0.2 μm and a purity of 99.98 mass%, and a powder of Al_2O_3 having an average grain diameter of 0.15 μm and a purity of 99.99 mass% were used. They were weighed such that the molar ratio of MgO to
20 Al_2O_3 became 50:50. Methanol as a dispersion medium, the powders of the raw materials, and nylon balls were placed in a resin-made ball mill vessel, and mixed (wet mixing) for 15 hours to yield a slurry. The slurry was dried (drying step) using a rotary evaporator, and the obtained dried powder was heat-treated at 1000°C in the atmosphere (heat treatment step) to yield a heat-treated powder. The heat-treated powder was dry-milled and then put into a resin-made ball
25 mill vessel together with methanol as a dispersion medium, the dry-milled heat-treated powder, and alumina balls, and wet-milling (milling step) was carried out to yield a slurry (raw material mixing step). The slurry was dried using a rotary evaporator, and the obtained dried powder was crushed and granulated to yield a granulated powder (drying and granulating step).

The granulated powder was HP-sintered in an Ar atmosphere at 1500°C, and under a pressure of 20 MPa to yield an HP sintered body. The HP sintered body was further HIP-sintered in an Ar atmosphere, in a temperature range of 1400°C to 1550°C, and under a pressure of 100 MPa to yield an HIP sintered body (sintering step). Since the HIP sintered body is reduced by the sintering in an inert gas atmosphere, an oxidation treatment was performed in an oxygen-containing atmosphere at 1500°C under normal pressure for 5 hours to yield a sintered body. The sintered body was processed into a desired shape (outline machining step), and the obtained sample was used for an evaluation.

(1-2) Examples 2, and 3

Each sample was prepared by the same steps as in Example 1 except that the molar ratio of MgO:Al₂O₃ was changed to 40:60 or 30:70, and subjected to an evaluation.

(1-3) Example 4

The same raw material powders as in Example 1 were weighed such that the molar ratio of MgO:Al₂O₃ became 20:80. The same wet mixing as in Example 1 was performed to yield a slurry (raw material mixing step). The slurry was dried using a rotary evaporator to obtain a dried powder. The dried powder was crushed and granulated without a heat treatment to yield a granulated powder (drying and granulating step). The granulated powder was molded at a pressure of 100 MPa (molding step), and then sintered in the atmosphere at 1750°C under normal pressure for 3 hours to yield a sintered body (sintering step). The sintered body was processed into a desired shape (outline machining step), and the obtained sample was subjected to an evaluation.

(1-4) Examples 5 to 7

Each sample was prepared by the same steps as in Example 4 except that the molar ratio of MgO:Al₂O₃ was changed to 10:90, 60:40, or 70:30, and subjected to an evaluation.

(1-5) Example 8

The same raw material powders as in Example 1 were weighed such that the molar ratio of MgO:Al₂O₃ became 50:50. The same wet mixing as in Example 1 was performed to yield a slurry. The slurry was dried using a rotary evaporator to yield a dried powder. The dried powder was heat-treated at 1000°C in the atmosphere to obtain a heat-treated powder. The

heat-treated powder was used as a granulated powder as it was without crushing. The granulated powder was subjected to the same HP sintering and HIP sintering as in Example 1 to yield a sintered body. The sintered body was processed into a desired shape, and the obtained sample was subjected to an evaluation.

5 (1-6) Example 9

A sample was prepared by the same steps as in Example 4 except that the molar ratio of $\text{MgO}:\text{Al}_2\text{O}_3$ was changed to 30:70, and subjected to an evaluation.

(1-7) Example 10

10 The same raw material mixing step (including the drying step, the heat treatment step, and the milling step) and drying and granulating step as in Example 1 were performed to yield a granulated powder. The granulated powder was subjected to an oxidation treatment in an oxygen-containing atmosphere at 1700°C and normal pressure for 5 hours. The oxidation-treated powder was used as a granulated powder as it was without crushing, and subjected to the same sintering step (HP sintering, HIP sintering, and oxidation treatment) and outline machining
15 step as in Example 1. The obtained sample was evaluated.

(2) Evaluation method

The following items were evaluated on the obtained samples.

(2-1) Relative density

The density of a sample can be determined, for example, by the Archimedes method.
20 The relative density of the sample can be expressed by the ratio of the actually measured density of the sample to the theoretical density corresponding to the composition of the sample (the total of the products of the respective theoretical densities of crystal phases constituting the sample and the respective volume ratios).

(2-2) Constituent phase, spinel phase volume ratio, spinel phase lattice constant, and spinel phase
25 composition

The crystal phase (constituent phase) constituting a sample, the volume ratio of the constituent spinel phase, and the lattice constant of the spinel phase can be determined from an X-ray diffraction pattern. The volume ratio and the lattice constant of the spinel phase are calculated using the Rietveld analysis software (TOPAS, manufactured by Bruker AXS GmbH)

from the X-ray diffraction pattern obtained using an XRD apparatus (D8 ADVANCE manufactured by Bruker AXS GmbH). Further, x of the composition $\text{Mg}_x\text{Al}_{2-2x}\text{O}_{3-2x}$ of the spinel phase is determined from the lattice constant.

(2-3) Average grain diameter, and D90/D10

5 The grain diameter of a crystal grain constituting a sample can be determined by an image analysis of a crystal grain in a mirror polished surface of the sample. That is, as the grain diameter of a crystal grain, a Heywood equivalent diameter that is obtained by converting the area of a crystal grain in the mirror polished surface into a circle is used. The grain diameters of 200 crystal grains are measured, and the value (D50) of the grain diameter distribution at 50%
10 (based on number) is used as the average grain diameter. Similarly, as D10 and D90, the values of the grain diameter distribution at 10% and 90% are used.

(2-4) Whiteness

As Whiteness of a sample, L^* in the system of CIE 1976 (L^* , a^* , b^*) is used. Whiteness value closer to 100 means closer to pure white. A sample having a thickness of 2 mm is
15 measured by a reflectance method (JIS Z-8722) using a colorimeter (ZE 6000, manufactured by Nippon Denshoku Industries Co., Ltd.), and the value calculated therefrom is used.

(2-5) Light transmittance

With respect to the light transmittance of a sample, the total light transmittance of a sample having a thickness of 2 mm is measured with an spectrophotometer (V-670 manufactured
20 by JASCO) using an integrating sphere, and the average light transmittance in an wavelength range of 400 to 800 nm is used.

(2-6) Dielectric loss

The dielectric loss of a sample can be determined by a waveguide method. The quality factor Q value is measured by placing the sample in a waveguide and using a network analyzer
25 (8720ES, Agilent Technologies, Inc.) at a measurement frequency of 10 GHz. At this time, an $f \cdot Q$ value (GHz) that is the product of the measurement frequency f (GHz) and the Q value is used as an index of the dielectric loss. A larger $f \cdot Q$ value means that the dielectric loss is smaller.

(2-7) Strength

As the strength of a sample, the three-point bending strength (JIS R1601) is used.

(2-8) Film formation test

The obtained sample is bonded with a Cu backing plate using In, and placed in a sputtering apparatus (CSL, manufactured by ULVAC, Inc.) as the sputtering target, and is

5 observed whether abnormal electrical discharge occurs or not.

(2-9) Unavoidable impurities

An inductively coupled plasma (ICP) analysis is performed with respect to the concentration of unavoidable impurities contained in a sample.

(3) Evaluation results

10 The evaluation results are shown in Table 1.

[Table 1]

Example	Overall composition MgO:Al ₂ O ₃ (molar ratio)	Heat treatment before granulation	Sintering method	Relative density (mass%)	Constituent phase	Volume ratio of spinel phase (%)	Lattice constant of spinel phase (Å)	Composition of spinel phase x	Average grain diameter (μm)	D90/D10	Whiteness L*	Light transmittance (%)	f-Q (GHz)	Strength (MPa)	Film formation properties
1	50:50	Yes	HP, HIP	99.8	Spinel	100	8.083	0.5	2.4	3.6	66	45	80000	345	○
2	40:60	Yes	HP, HIP	99.6	Spinel	100	8.037	0.4	2.3	2.4	75	40	74000	335	○
3	30:70	Yes	HP, HIP	99.6	Spinel	100	8.000	0.3	2.1	2.7	73	42	89000	320	○
4	20:80	No	Normal pressure	99.9	Spinel, Al ₂ O ₃	36	8.073	0.48	5.3	2.8	82	27	107000	340	○
5	10:90	No	Normal pressure	100	Spinel, Al ₂ O ₃	16	8.076	0.48	6.5	2.2	82	27	96000	385	○
6	60:40	No	Normal pressure	99.5	Spinel, MgO	88	8.082	0.5	5.2	3.2	77	36	88000	355	○
7	70:30	No	Normal pressure	99.6	Spinel, MgO	73	8.083	0.5							○
8	50:50	Yes	HP, HIP	99.8	Spinel	100	8.083	0.5	-	-	30	1	48000	-	△
9	30:70	No	Normal pressure	96.5	Spinel	100	8.000	0.5	1.6	2.6	98	4	-	230	△
10	50:50	Yes	HP, HIP	99.8	Spinel	100	8.083	0.5	12						○

(3-1) Examples 1 to 3

In Examples 1 to 3, the relative densities were as high as 99.6 to 99.8 mass%. Further, with respect to the constituent phase, the volume ratio of the spinel phase was 100%, and the composition of the spinel phase was $x = 0.5$ to 0.3 ($\text{Mg}_{0.5-0.3}\text{Al}_{1-1.4}\text{O}_{2-2.4}$) reflecting a molar ratio of MgO to Al_2O_3 of the raw materials of 50:50 to 30:70. It is presumed that the above result was obtained because the reaction between the raw material powders was promoted by the heat treatment step before granulation. The average grain diameter (D50) of the sintered body was 2.1 to 2.4 μm , and the D90/D10 was 2.4 to 3.6. Whiteness was as high as 66 to 75 indicating a white color, and the light transmittance was 40 to 45%. Further, the f·Q value was 74000 to 89000 GHz indicating a low dielectric loss. The bending strength was as high as 320 to 345 MPa, which was sufficient as the strength for a sputtering target member. The abnormal electrical discharge occurred rarely in sputtering, and good film formation could be performed.

(3-2) Example 4 to 7

In Examples 4 to 7, the relative density was as high as 99.5 to 100 mass%. Further, with respect to the constituent phases, the volume ratio of the spinel phase was as low as 16 to 88%, in Examples 4 and 5, the Al_2O_3 phase was additionally observed, and in Examples 6 and 7, the MgO phase was observed. The composition of the spinel phase was $x = 0.48$ to 0.5 ($\text{Mg}_{0.48-0.5}\text{Al}_{1.04-1}\text{O}_{2.04-2}$). The average grain diameter (D50) of the sintered body was 5.2 to 6.5 μm , which was larger than that in Examples 1 to 3. It is considered that grains grew due to a high sintering temperature. The D90/D10 was 2.2 to 3.2. Whiteness was 77 to 82 indicating a white color, and the light transmittance was 27 to 36%, which was lower than that in Examples 1 to 3. The f·Q value was 88000 to 107000 GHz indicating a low dielectric loss, and the bending strength was 340 to 385 MPa, which was sufficient for a sputtering target member. The abnormal electrical discharge occurred rarely in sputtering, and good film formation could be performed.

(3-3) Example 8

In Example 8, the relative density was as high as 99.8 mass%, the volume ratio of the spinel phase was 100%, and the composition of the spinel phase was $x = 0.5$ ($\text{Mg}_{0.5}\text{AlO}_2$).

Whiteness was 30 indicating a black color, and the light transmittance was 1%. The f·Q value was 48000 GHz.

(3-4) Example 9

5 In Example 9, the relative density was 96.5 mass%, the spinel phase volume ratio was 100%, and the composition of the spinel phase was $x = 0.5$ ($\text{Mg}_{0.5}\text{AlO}_2$). The average grain diameter (D50) of the sintered body was 1.6 μm , and the D90/D10 was 2.6. Whiteness was 98 indicating a white color, the light transmittance was 1%, and the f·Q value was 48000 GHz.

(3-5) Example 10

10 In Example 10 the relative density was as high as 99.8 mass%, the volume ratio of the spinel phase was 100%, and the composition of the spinel phase was $x = 0.5$ ($\text{Mg}_{0.5}\text{AlO}_2$). The average grain diameter (D50) of the sintered body was as large as 12 μm .

(3-6) Unavoidable impurities

All the concentrations of unavoidable impurities in Examples 1 to 10 were from several ppm to below the lower limit of measurement.

15 Although the embodiment of the invention has been described above in detail, those skilled in the art will readily understand that various modifications may be made without substantively departing from the new matters and the effects of the invention. Therefore, such modifications are entirely included within the scope of the invention. For example, any term described at least once together with a broader or synonymous different term in the description
20 may be replaced by the different term at any place in the description. Moreover, the configuration or action of the target member for physical vapor deposition, the physically vapor-deposited film, the layered structure, etc. are not limited to those described in the present embodiment, but may include various modifications.

What is claimed is:

1. A target member for physical vapor deposition comprising Mg, M (M is a trivalent metal
5 element), and O as main components, wherein
a molar ratio of the Mg to the M in terms of the oxides of MgO and M₂O₃ respectively is
70:30 to 10:90.
2. A target member for physical vapor deposition comprising Mg, M (M is a trivalent metal
10 element), and O as main components, and comprising a crystal phase having a spinel structure.
3. The target member for physical vapor deposition according to claim 1 or 2, wherein the
M is one or two selected from the group consisting of Al and Ga.
- 15 4. The target member for physical vapor deposition according to any one of claims 1 to 3,
wherein a light transmittance for a thickness of 2 mm is 60% or less.
5. The target member for physical vapor deposition according to any one of claims 1 to 4,
wherein a dielectric loss at 10 GHz is 45000 GHz or more in terms of an f·Q value.
20
6. The target member for physical vapor deposition according to any one of claims 1 to 5,
wherein Whiteness is 30 or more.
7. A sputtering target member comprising the target member for physical vapor deposition
25 according to any one of claims 1 to 6.
8. A method for producing a physically vapor-deposited film, comprising physically vapor
depositing the physically vapor-deposited film on a underlayer using the target member for
physical vapor deposition according to any one of claims 1 to 6.

9. A method for producing a layered structure, comprising:
physically vapor depositing a physically vapor-deposited film on a underlayer using the
target member for physical vapor deposition according to any one of claims 1 to 6, and
5 forming a ferromagnetic layer on the physically vapor-deposited film, wherein
the underlayer is a ferromagnetic layer, and the physically vapor-deposited film is a
tunnel barrier layer.