Soft Magnetic Properties of CoNbZr amorphous Films with Pd addition

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The present paper is to investigate the phase stability and soft magnetic properties of amorphous CoNbZr films when Pd is added as a substitution for CoNbZr alloys. The films were prepared by a RF magnetron sputtering method. The CoNbZrPd films deposited on Si wafers exhibited amorphous structures being independent upon the amount of Pd added in the films. On the addition of 4.34% Pd, the excellent soft magnetic characteristics of the films were observed with a coercive force of 0.54 Oe and an anisotropy field of 11 Oe, whereas a coercive force of 1 Oe and an anisotropy field of 3.5 Oe were shown in the film without the addition of Pd. The increased anisotropy field and low coercive force of the films may be attributed to the occupancy of Pd in the preferred sites parallel to the external magnetic field applied on the deposition process. A permeability of about 1100 was kept constant in the operation frequency ranging up to 100 MHz, which can be explained by the Landau-Lifshitz formula.

1. INTRODUCTION

Frequency dependence of magnetic characteristics[1,2] such as permeability and loss factors for the magnetic materials can be expected by the formula[3] of a magnetic resonance form derived from the Landau-Lifshits equation and classical eddy current equation. The formula shows that the natural resonance frequency has a linear relation to the saturation magnetization M_s and anisotropy field H_k, and eddy current loss is inversely dependent on electrical resistivity ρ. Consequently, the materials must possess high saturation magnetization, electrical resistivity and anisotropy field for high frequency applications. Among many material systems investigated so far, CoAlO- and FeAlO-based films with addition of Pd[4,5] are considered to be the proper candidates in that Pd occupies preferentially the specific axis of the unite cell and then induce new crystal structure (bct and fct) resulting in high magnetic anisotropy. Despite the advantage of the high anisotropy, the magnetic materials show a high coercive force of more than 1 Oe, a saturation magnetization of 0.9 T and a low permeability of 200. On the other hand, CoNbZr amorphous films are well known to have relatively high magnetization and low coercivity but low anisotropy due to locally inhomogeneous compositional and structural state. If such a local inhomogeneity can be minimized by adding other elements like Pd, anisotropy and operating frequency range are expected to increase.[6]

This study presents the magnetic properties and structural change in CoNbZr amorphous films containing Pd as a partial substitute for Co which can be a new soft magnetic material with high permeability in the range of MHz to GHz.

2. EXPERIMENTAL PROCEDURES

2.1 Sample preparation

CoNbZrPd amorphous films were fabricated on Si (100) wafer in a way that the materials were sputtered in a target geometry which Pd chips ($5\times5\times1$ mm^3) were kept placed on the circular $Co_{84}Nb_{12.5}Zr_{3.5}$ plate. The initial vacuum is maintained below 5×10^{-7} torr. Then, Ar gas of 8cc/min was flowed into the chamber and the input power of 200W was charged between two electrodes under a pressure of 2 mtorr. Pd amount of the film composition was varied by deposition under the geometry which is of changing the number of the chips on the plate. The films deposited on the Si wafers are in 1.3μm thick circular shape of 5mm diameter.

2.2 Characterization

The thickness of CoNbZr-Pd film was measured by a surface profiler(Tencor). The structural analysis and chemical composition identification of the CoNbZr-Pd films were carried out in X-ray diffraction experiments and an electron probe micro analysis. Since the films prepared in this study are amorphous state and have no the notation of crystallographic orientation, the measurement of magnetic properties was carried out by application of external magnetic field in all possible in-plane directions. The magnetically easy and hard axes were arbitrarily chosen from the finding of maximum and minimum D.C. permeabilities taken in the magnetization M versus external magnetic field H curves. The saturation magnetization and the coercive force were obtained at an applied magnetic field of 50 Oe using a vibrating sample magnetometer. The magnetic anisotropic field H_k was determined by comparing the coercive forces observed in the M-H curves under magnetic field application along magnetically easy axis and hard axis. The permeabilities were measured in the frequency ranging MHz to GHz by detecting the voltage signal which is transmitted from a one turn coil jig-circuit system to a network analyzer (HP 8752C).[7] The measured peremability was compared with the calculated by the formulas derived from the Landau-Lishiftz kinetics.

3. RESULT AND DISCUSSION

3.1 Deposition and Structure

In the deposition process, there are several methods of controlling chemical composition of the films and the most common deposition is to have materials sputtered from the target geometry where the number of the element chips putting on the other element plate is changed. Except for the target geometrical structure, the other deposition variables like substrate temperature, input power and Ar gas pressure were the same for the film fabrication process so as to control the Pd amount existing in the properties which is in association with not only intrinsic structural nature, short range ordering of the amorphous phase in the present study, but also extrinsic film nature such as size and shape and surface roughness. Among the various factors influencing the magnetic characteristics, chemical compositional change and its accompanying structural modification are key factors in the amorphous film containing Pd element. The chemical compositional analysis of the films made in the deposition under the target geometrical conditions is shown in the Fig. 1. The Pd amount of chemical composition increases in proportion to the number of the chip and Co amount decreases linearly with the increase of Pd. Nb and Zr showed to be constant in chemical composition of the films irrespective of change in the number of the Pd chip. Considering that Pd is similar with Co in atomic radii and the two elements can mix completely each other, the result in the Fig. 1 represent that Pd can substitute with Co in the short range ordering forms of amorphous structure. Fig. 2 represents X-ray diffraction patterns of $Co_{91.50}Nb_{6.80}Zr_{1.70}$-Pd films, deposited at an input power of 200W and an argon pressure of 2 mtorr, on the Si substrate. As seen, all films done in this study shows the halo pattern in the pattern, independent on the amount of Pd substituted for Co, which indicates the existence of amorphous state. From the diffraction pattern, the peak position of amorphous phase and the broadening of peaks were found to decrease as the Pd concentration increases, as seen in the right side of the Fig. 2.

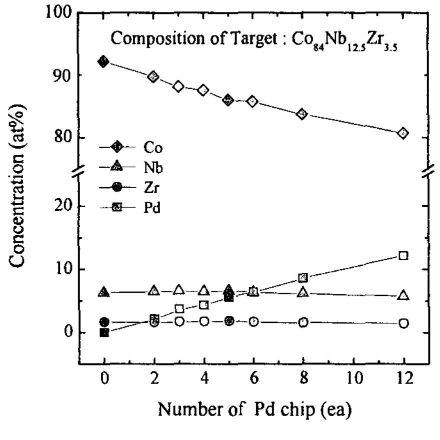

Fig. 1. Changes in the composition as a function of number of Pd chips for CoNbZr-Pd films.

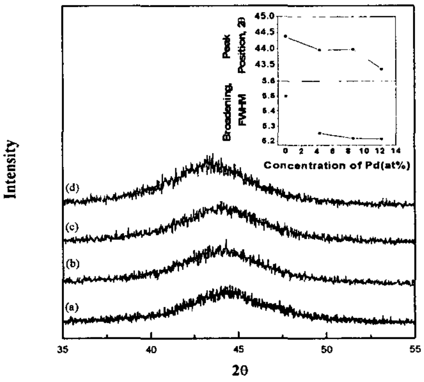

Fig. 2. X-ray diffraction patterns of as-deposited CoNbZr-Pd films
(a) $Co_{91.50}Nb_{6.80}Zr_{1.70}$ (b) $Co_{87.56}Nb_{6.45}Zr_{1.65}Pd_{4.34}$
(c) $Co_{83.73}Nb_{6.22}Zr_{1.45}Pd_{8.6}$ (d) $Co_{80.26}Nb_{6.13}Zr_{1.40}Pd_{12.21}$

The peak position shift represents the change in the average neighboring atomic distance in the amorphous phase and the broadening indicates the degree of irregularity in amorphous state. Based on the indication of peak position and broadening, Pd addition to the film was thought to give the expansion of short-range ordering form and the reduction of local fluctuation.

3.2 Magnetic properties

Fig. 3 shows the variation of saturation magnetization and coercive force with Pd amount for the films. As seen in the Fig. 3 (a), saturation magnetization decreases with increasing Pd amount added in the film, which may result from the reduction of ferromagnetic element Co composition along with increase of Pd. The values of saturation magnetization are above 1.1T. Fig. 3(b) represents the variation of coercive force with Pd amount for the films. The $Co_{91.50}Nb_{6.80}Zr_{1.70}$ film without Pd addition has almost a coercive force of 1 Oe. The lowest coercive force 0.54 Oe was obtained in the CoNbZr film containing 4.34 at% Pd and the coercive force increase to reach more than 1Oe as Pd is added over 6.2 at%. All films showed that the coercive force at the applied field along the easy axis is slightly smaller than that under applied field along the hard axis. The decrease in coercivity in the range of Pd up to 4.34at% may be caused by the combined effect of the expansion of short-range ordering forms mentioned above and the magnetic ordering of amorphous phase induced by the external magnetic field on the deposition. On the addition of Pd over 6at%, the internal stress due to the local structural fluctuation described in the Fig. 2 allows the retard of magnetic domain wall motion and magnetization rotation which may induce the increase of the coercivity.

Fig. 4 shows the anisotropy field H_k of the CoNbZr-Pd film with respect to Pd addition. In general, magnetic anisotropy magnetic body is an energy form representing the resistance to rotatie magnetic moment from the specific direction, which consists of film. The Pd addition to the CoNbZr film influences the magnetics of magnetocrystalline anisotropy, anisotropy induced by external magnetic field and anisotropy due to magnetostriction. However, the CoNbZrPd films don't have magnetocrystalline anisotropy and the small magnetostriction can be negligible as compared with the induced anisotropy. The observed anisotropy of the films may be caused by the preferentially alignment of ferromagnetic element Co parallel to the applied field over Pd in short range ordering forms on deposition. The anisotropy was increased with increasing Pd amount added in the film and it is above 10Oe in the range of Pd 2~5 at%. When Pd is added with more than 7at%, the anisotropy field is lower than that of the film without Pd addition.

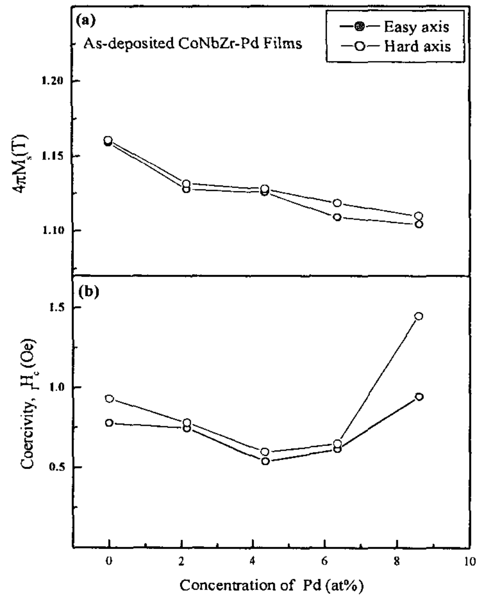

Fig. 3. Changes in the saturation magnetization (a) and the coercivity (b) as a function of amount of Pd addition for amorphous CoNbZr films

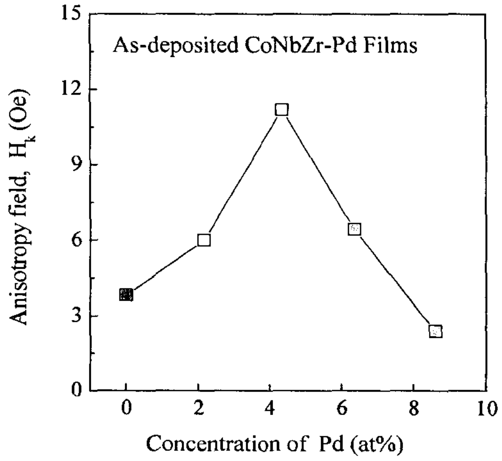

Fig. 4. Change in the anisotropy field as a function of amount of Pd added in the amorphous CoNbZr films.

Fig. 5 shows the frequency dependence of permeability for the $Co_{87.56}Nb_{6.45}Zr_{1.65}Pd_{4.34}$ film. The film exhibits the constant permeability 1100 as the frequency reaches up to 100 MHz, whilst the CoNbZr film has approximately a permeability of 750. The frequency dependence of magnetic film permeability can be related to the eddy current loss and the magnetic anisotropy dispersion in the film. The one is the permeability-frequency equation under consideration of magnetic loss due to eddy current behavior.

$$\mu' = (\frac{\mu_i^{dc}}{K})[\frac{\sinh K + \sin K}{\cosh K + \cos K}]$$
$$\mu'' = (\frac{\mu_i^{dc}}{K})[\frac{\sinh K - \sin K}{\cosh K + \cos K}] \quad with \quad K = 2.0\pi t_m \sqrt{\frac{\mu_i^{dc} f}{\rho}} \qquad \cdots\cdots\cdots\cdots (1)$$

μ', μ'' and μ_i^{dc} are real part, imaginary part of permeability and initial permeability in d.c. field. t_m, f, ρ are the film thickness (1.3μm), operation frequency, electrical resistivity (134μΩ·cm), respectively. The other shows the relation of permeability to operation frequency on a basis of magnetic anisotropy dispersion in amorphous phase.

The form expressed by Landau-Lifshitz kinetic formula presents a linear equation[8,9] showing the relation of permeability to frequency, under the assumptions that (1) anisotropy field H_k is far lower than saturation magnetization M_s, (2) damping constant α is far below 1, (3) external field H_{ex} is applied to magnetically hard axis and is lower than anisotropy field H_k of film and (4) the film is very thin so as to neglect the eddy current effect.

$$\mu_{xx} = \frac{\gamma M_S}{(\gamma H_k + i\alpha\omega)} \times [1 + \frac{\omega^2(1-\alpha^2)^2}{(\gamma H_k + \gamma M_S + i\alpha\omega)(\gamma H_k + i\alpha\omega) - \omega^2}] + 1 \quad \text{.............................} \quad (2)$$

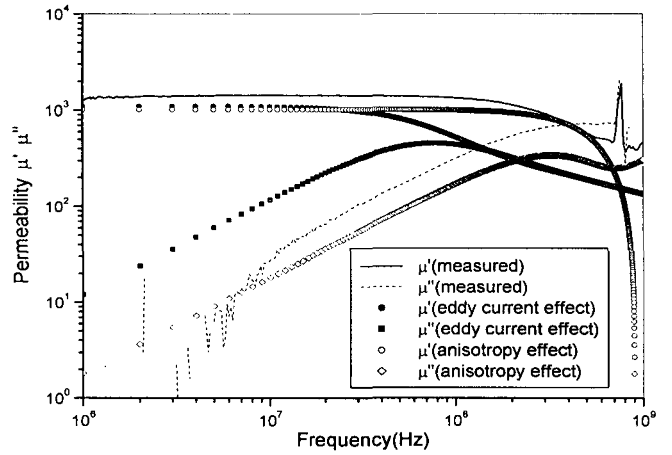

Fig. 5. Permeability vs. frequency for the $Co_{87.56}Nb_{6.45}Zr_{1.65}Pd_{4.34}$

where μ_{xx} is in-plane permeability of the film, γ gyro-constant (2×10^5 m/A·s), α damping constant (0.05) and $\omega(=2\pi f)$ angular frequency. These equations (1), (2) and the results of saturation magnetization vs. Pd amount (see Fig. 5) represent that the films containing Pd 2~5at% can exhibit high permeability and high resonance frequency. These measured permeabilities agree fairly well with the calculated value using equation (1) in the frequency ranging up to 50MHz and with using equation (2) in the range of 50MHz to 1GHz. These results state that the magnetic domain wall motion contribute to the magnetization up to 50MHz. In the frequency range of several hundred MHz to GHz, the rotation process can be a major mechanism of magnetization and the anisotropy field can affect the change of permeability in the high frequency range. The improved permeability is caused by the enhanced anisotropy field of the film with Pd addition.

4. Conclusions

CoNbZr-Pd amorphous films were fabricated on Si wafers and their magnetic properties were investigated. All films have amorphous state which is independent upon the amount of Pd added in the samples. The films containing Pd 4.34at% showed high anisotropy field and low coercive force which induce higher permeability in the frequency of several hundred MHz, as compared with the film without Pd addition..

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