様　式　C－１９、F－１９－１、Z－１９　（共通）

科研費 KAKENHI

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研究課題名（和文）粒子傾斜構造によるトンネル磁気-誘電効果薄膜の広帯域・高周波化

研究課題名（英文）Development of wide-frequency band and high-frequency tunneling magneto-dielectric effect in gradient granular structure

研究代表者

曹　洋（Cao, Yang）

東北大学・学際科学フロンティア研究所・助教

研究者番号：５０８０４５９８

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研究成果の概要（和文）：2014年に私たちのグループによって発見されたナノコンポジットのトンネリング磁気誘電（TMD）効果は、最大誘電率変動の狭いピークを特徴としています。 これにより、広い周波数範囲での磁電アプリケーションが制限されます。 実用的な観点から、広帯域TMD応答の開発が強く求められています。 この研究では、広帯域・高周波化TMD効果を示すことができる粒子傾斜構造を提案および製造しました。 さらに、従来のナノグラニュラーフィルムに少量のSiを添加するだけで、最大8.5%という前例のない高いTMD効果を実現しました。

研究成果の学術的意義や社会的意義
This research results may have potential in practical high-frequency band device applications in such as 5G cell phones as well as other high-frequency mobile devices.

研究成果の概要（英文）：Tunnelling magneto-dielectric (TMD) effect in nanocomposites discovered by our group in 2014 features a narrow peak of the maximum dielectric variations. This limits its magnetoelectric application over a wide frequency range. From the practical viewpoint, it is highly demanded to develop the broadband TMD response. In this research, I have proposed and fabricated a composition-gradient multilayer nanogranular structure that can exhibit a broadband TMD effect up to megahertz frequency range. Additionally, I have realized the unprecedented high TMD response of up to 8.5% with small addition of Si into the conventional nanogranular films.

研究分野： Magnetic and dielectric films

キーワード： composition-gradient　broadband frequency　magneto-dielectric　Si dopant　dielectric enhanment

１．研究開始当初の背景

Magnetic nanogranular structures that comprise nanometer-sized magnetic granules dispersed within insulating or metallic host matrices represent a robust platform for fundamental studies of disordered solids because of their adjustable functionality, including their electronic, magnetic, and optical properties.[1] Depending on the different granule/matrix ratio used, these structures with relatively high granular content can host a variety of intriguing properties, including high-frequency soft magnetic properties,[2] tunneling magnetoresistance[3] and the giant Faraday effect.[4]

When the magnetic granular content is reduced further, the resulting structures may fall into the dielectric regime. From this perspective, the tunneling magneto-dielectric (TMD) effect in insulating nanogranular films was recently reported by our group.[5] Upon application of a magnetic field (H), the dielectric permittivity (ε'_0) of the nanogranular films increases, where the increased value is denoted by ε'_H, over the frequency range of the AC electric field; the increase in the dielectric permittivity, i.e., the TMD ratio denoted as $\Delta\varepsilon'/\varepsilon'_0$, is caused by spin-dependent charge oscillation via interaction with the insulator matrix between the magnetic granules. The TMD effect features a small TMD ratio in both the low- and high-frequency ends and. However, the magnitude of TMD ratio beyond the f_{TMD} frequency range is almost negligibly small, which restricts the application of the TMD effect over a broad frequency range. On the other hand, the achieved TMD response so still remains unsatisfactory, it is high demanded to further enhance the TMD ratio.

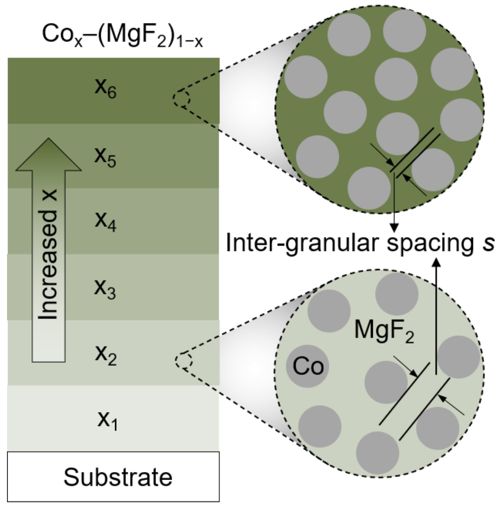

Fig. 1. Schematic of the configuration of the CGM Co_x-$(MgF_2)_{1-x}$ nanogranular structure with increasing Co granular content (x). The right side presents a detailed depiction of the dispersed state of the Co granules in the MgF_2 matrix with different inter-granular spacing (s) in the different layers. The granule size variation in the different layers was ignored here. This figure is copied from Ref [6].

２．研究の目的

Taking inspiration from Functionally Graded Materials (FGM) that show a gradual composition variation, we present a proof-of-concept study of use of a Composition-Graded Multilayer (CGM) nanogranular structure, as depicted schematically in Fig. 1, to achieve a broadband TMD effect. The CGM film structure is constituted by an upwardly-graded increase in the Co granular content from x_1 to x_6 in the different compositional layers, which means that the average inter-granular spacing may undergo a small step-varied reduction on the sub-nanometer scale, provided that the variations in the granule size over the different layers are ignored.

３．研究の方法

The compositional grading of the Co_x-$(MgF_2)_{1-x}$ films was realized by magnetron co-sputtering of Co and MgF_2 targets on $Si/SiO_2/Ti/Pt$ substrates under an Ar gas pressure of 0.5 Pa at room temperature. Two target sources are located in one chamber with an angle of 45° relative to the substrates. The Co content of each layer was carefully regulated by increasing the input power from 80 to 120 W at intervals of 10 W with fixed sputtering power of MgF_2 target to 150 W. The substrate was rotated at a speed of 10 rpm to achieve a uniform granular state. Structures were observed using a field-emission transmission electron microscopy and a Cs-corrected 200 kV high-angle annular dark-field (HAADF)-type scanning transmission electron microscope (STEM). The magnetic behavior was measured using a vibrating sample magnetometer. The dielectric and magneto-dielectric properties were measured using an inductance-capacitance-resistance meter within a 1-1000 kHz frequency range and an impedance analyzer in the range of 1 kHz-100 MHz, with magnetic fields ranging up to ±10 kOe.

４．研究成果

成果 1:

To achieve the stoichiometry composition control of Co content in each layer, the deposition rates of both Co and MgF_2 targets have been calibrated. The CGM structures consisted of step-varied compositions with corresponding increases in their Co granular content from 0.23 to 0.26.[6] The thickness of each single layer was fixed at 40 nm by controlling the sputtering duration. For comparison, conventional uniform nanogranular films composed of Co_x-$(MgF_2)_{1-x}$ with different x values were also deposited. The TMD response for uniform granular films is very small on the low-frequency side near 100 kHz and it also

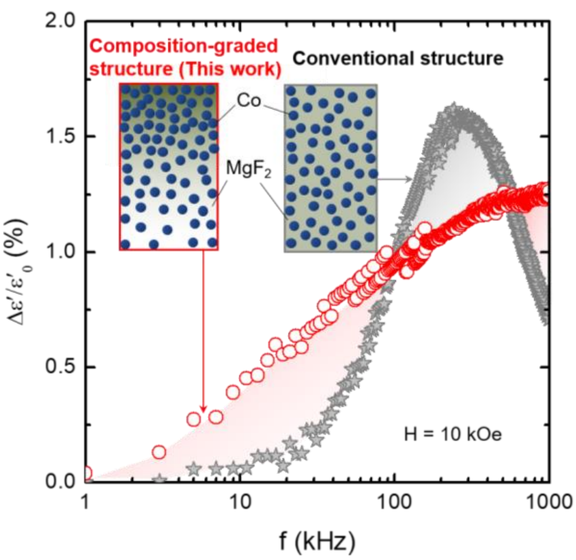

Fig. 2. Frequency dependence of the dielectric variations ($\Delta\varepsilon'/\varepsilon'_0$) under an applied magnetic field H = 10 kOe. Inset: the schematic depiction of the composition-graded structure and the conventional nanogranular structure.

decreased dramatically at the high-frequency side, despite the fact that there is a peak TMD ratio of 1.6% at 300 kHz as shown in Fig. 2, which is one of the TMD effect characteristics that has been confirmed in various nanogranular material systems with different matrices, such as AlF_3. In contrast, the TMD response for CGM shows a fairly flat peak and retains a relatively high TMD ratio in both the low-frequency range and the high-frequency range. Even at higher frequency range, it can still remain as high as 0.53% at 10 MHz, which is an almost 9-fold enhancement in comparison with that for conventional one (0.06%).

成果 2:

We report a large enhancement of the tunneling magneto-dielectric (TMD) effect in Co–MgF_2 granular films induced by doping using a small amount of Si.[7] The Co-MgF_2-Si granular nanocomposites were deposited on Si/SiO_2/Ti/Pt substrates using a triple-source radio-frequency magnetron sputtering technique using Co and Si disk targets and a MgF_2 compacted powder target to realize precise composition control. This minor addition of Si is dispersed uniformly in the MgF_2 matrix and acts by inhibiting the interdiffusion between the Co and MgF_2 phases, thus enhancing the magnetization when compared with the case of the corresponding undoped Co–MgF_2 films; this consequently results in a greatly enhanced peak dielectric variation (TMD ratio, $\Delta\varepsilon'/\varepsilon'_0$), as indicated by theoretical fittings. Extension of this Si doping effect to CoFe–MgF_2 films led to record-high $\Delta\varepsilon'/\varepsilon'_0$ of 4.3% at 10 kHz and 8.5% at 200 kHz under application of a magnetic field (H) of 10 kOe, while remaining as high as 2.1% even under H = 1 kOe. This study presents a simple but highly effective approach to enhance the TMD effect in granular nanocomposites, thus opening up the prospect of development of high-performance magnetoelectric devices.

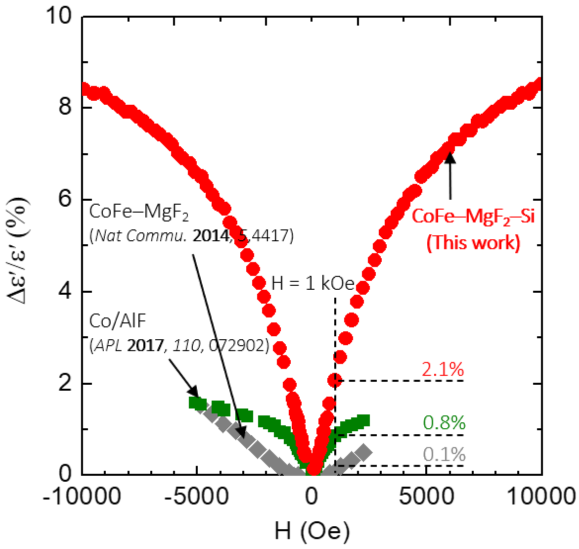

Fig. 2. Frequency dependence of the dielectric variations ($\Delta\varepsilon'/\varepsilon'_0$) under an applied magnetic field H = 10 kOe. Inset: the schematic depiction of the composition-graded structure and the conventional nanogranular structure.

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５．主な発表論文等

〔雑誌論文〕　計0件

〔学会発表〕　計3件（うち招待講演　1件／うち国際学会　3件）

１．発表者名 Yang Cao
２．発表標題 Tunneling Magneto-Dielectric (TMD) effect: Recent advances and future perspectives
３．学会等名 The 10th Pacific Rim International Conference on Advanced Materials and Processing (PRICM10)（招待講演）（国際学会）
４．発表年 2019年

１．発表者名 Yang Cao
２．発表標題 Emerging Tunneling Magneto-Dielectric (TMD) Effect and Recent Progress
３．学会等名 The 11th China-Japan Symposium on Ferroelectric Materials and Their Applications（国際学会）
４．発表年 2019年

１．発表者名 Yang Cao
２．発表標題 Structure and Magneto-dielectric Properties in Co-F-C Nanocomposites
３．学会等名 The 13th Pacific Rim Conference of Ceramic Societies (PACRIM13)（国際学会）
４．発表年 2019年

〔図書〕　計0件

〔産業財産権〕

〔その他〕

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６．研究組織

	氏名 （ローマ字氏名） （研究者番号）	所属研究機関・部局・職 （機関番号）	備考

７．科研費を使用して開催した国際研究集会

〔国際研究集会〕　計0件

８．本研究に関連して実施した国際共同研究の実施状況

共同研究相手国	相手方研究機関