

## Structure and Properties of Diamond Like Carbon-Magnetic Metal Nano-composite Films

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### ABSTRACT

As the advent of internet of things, multi-functional devices with environment-friendly property and low energy consumption is required. In this project, nano-composite films with diamond like carbon (DLC) matrix-magnetic metal particles combination will be discussed. The DLC-Co nano-composite films show nano-composite structure with clear interface between the amorphous C matrix and Co particles. From X-ray diffraction, the DLC-Co films have a amorphous structure. The magnetic properties of these films change from ferromagnetic character to superpara-magnetic character by controingll methane flow rate.

### 1. Introduction

As developments of micro-electric devices, it requires multi-functional materials urgently, such as magnetic-dielectric films. In our previous work, it has found that the nano-composite films show large dielectric properties and high-frequency magnetic properties[1,2]. We have proved that the insulation property and superparamagnetic property are important to obtain the magnetic-dielectric properties, which require uniform dispersion of magnetic metal particles in insulator phase. However, the resistivity of the films is too low for the magnetic-electrical properties, such as magnetoresistance and dielectric response properties. Nano-composite films with diamond-like carbon (DLC) -Co metal particles will be a good candidate for this purpos. Diamond-like Carbon (DLC) film with high resistivity, low friction coefficient, excellent corrosion resistance and biocompatibility is thought to be ideal insulation material. It is a three-dimensional cross-network structure formed by sp<sup>3</sup> hybrid bond of Diamond phase and sp<sup>2</sup> hybrid bond of graphite phase, containing a small amount of hydrogen atoms. Strongly varying microstructure and chemical composition of DLC films are found though the selection of deposition methods and the regulation of preparation conditions. Plasma enhanced chemical vapor deposition (PECVD) has great advantage of synthesizing density DLC film[3]. Moreover, it can be changed from metallic to insulating by carbon structure controlling. In this research, DLC-Co nano-composite films have been produced by hybrid PECVD-sputtering method. The resistivity of the films has been controlled by the gas flow rate. The structure, electrical and magnetic properties of DLC-Co films have been investigated.

### 2. Method

The DLC-Co nano-composite films were deposited on Si (100), Pt/Si and quartz substrates by hybrid PECVD-sputtering method, which consists of chemical vapor deposition (CVD) using methane (CH<sub>4</sub>)/argon gas mixture as a precursor and co-sputtering of a metal Co target. Flow rate of CH<sub>4</sub> of 3, 6, 7.5, 9 sccm included to chamber was changed to control carbon content. The structure of the as-deposited DLC-Co films was analyzed by X-ray diffraction (XRD, Bruker NEW D8

ADVANCE) using Cu K $\alpha$  radiation and field-emission transmission electron microscopy (FETEM, Hitachi 4300E). The composition of the films were identified by EDX (Ultra Dry, Thermo Fisher Scientific Inc.). Film thickness was measured by surface profile-meter (Dektak 8). Electrical resistivity was measured by a conventional four-point probe method. The magnetization curves were identified by a vibrating sample magnetometer (VSM, BHV-30SS, Rikendensi). All the measurements reported in this paper were carried out at room temperature.

### 3. Results and Discussion

The structure of DLC-Co film (typical sample) has been investigated by the XRD. As shown in Fig. 1, XRD of DLC-Co films illustrates the all the films show amorphous structure. From 3 sccm to 9sccm, there is no obvious peak can be found for C, which indicate the Carbon is amorphous. Whereas, from HRTEM observation, the Co particles should have nano-crystalline structure. The Co particles have a very well dispersion in the Carbon matrix. These DLC-Co films show different microstructure, which will lead to different electric and magnetic properties. With the flow rate of CH<sub>4</sub> increasing from 3-9 sccm, the Co content of DLC-Co films decrease, which will induce the various magnetic properties.

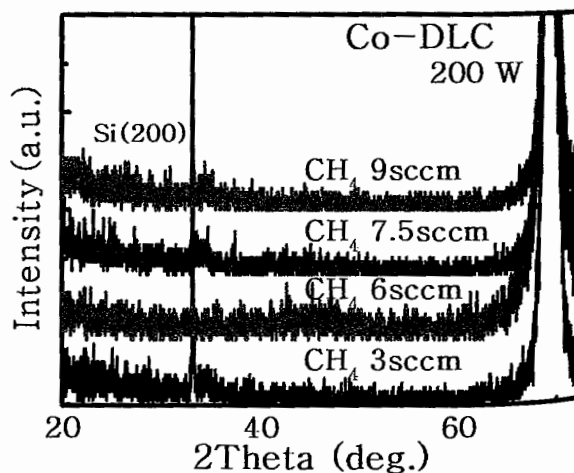


Fig. 1. XRD of the DLC-Co nano-composite film.

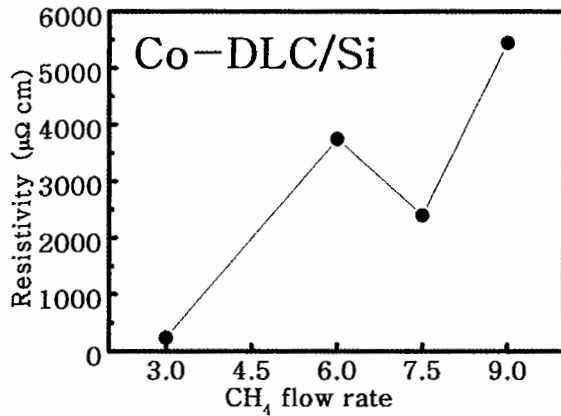


Fig. 2. Resistivity of DLC-Co nano-composite film.

Fig. 2 shows that the resistivity  $\rho$  increases from 200 to 5500  $\mu\Omega\cdot\text{cm}$  by increasing the  $\text{CH}_4$  flow rate from 3 to 9 sccm, which can be explained by Co content change.

The magnetic properties of DLC-Co films with different  $\text{CH}_4$  flow rate are shown at Fig. 3. The saturation magnetization ( $B$ ) of films increases from 200 Gs to 4600 Gs, with the  $\text{CH}_4$  flow rate increasing. As shown in Fig. 3 (b), when  $\text{CH}_4$  flow rate is 3 sccm, the films show ferromagnetic properties, with the  $B$  of 4600 Gs. The coercivity of DLC-Co nano-composite films is 21 Oe, which could show a soft magnetic property and have potential for the high-frequency soft-magnetic properties. When the  $\text{CH}_4$  flow rate increase to 9 sccm, the films show superpara-magnetic properties. However, the magnetization is only 200 Gs. These films also show high resistivity. Therefore, this DLC-Co films could be studied as magnetoresistance material.

#### 4. Concluding Remarks

The DLC-Co nano-composite films have been produced by hybrid PECVD-sputtering method. All the films show amorphous microstructure. The  $\rho$  of C-Co films increases from 200 to 5500  $\mu\Omega\cdot\text{cm}$ , By increasing the  $\text{CH}_4$  flow rate from 3 to 9 sccm. When  $\text{CH}_4$  flow rate is 3 sccm, the films show ferromagnetic properties, with the  $B$  of 4600 Gs. The coercivity of DLC-Co nano-composite films is 21 Oe. When  $\text{CH}_4$  flow rate is 9 sccm, the films show superpara-magnetic properties. By controlling the gas flow rate, the resistivity increases, and the films change from ferro-magnetic films to superpara-magnetic films. It is noteworthy, the DLC-Co nano-composite films has the promising to realize both high resistivity and superpara-magnetic properties in the further work, which could be studied as magnetoresistance (MR) and dielectric response material.

#### Acknowledgements

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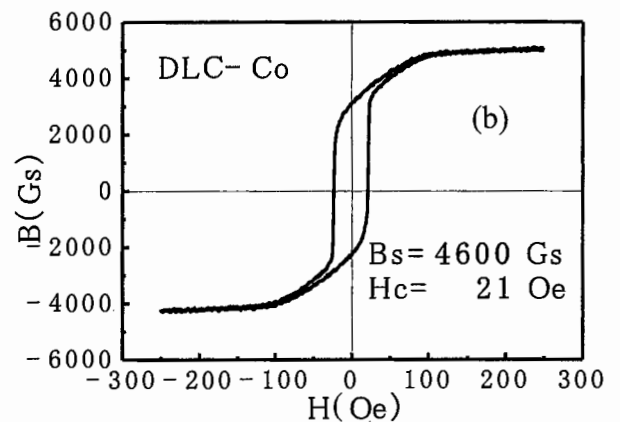
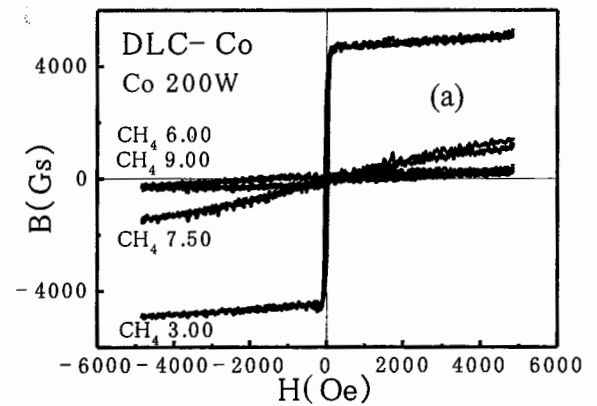


Fig. 3. Magnetization of DLC-Co nano-composite film.

#### References

- [1] Y. W. Zhang, S. Ohnuma, H. Masumoto, *IEEE Transactions on Magnetics*, 47 (2011) 3795-3798.
- [2] Y. W. Zhang, N. Kobayashi, S. Ohnuma, M. Nose, and H. Masumoto, *Journal of Magnetism and Magnetic Materials*, 401 (2016) 925-928.
- [3] T. Takeno, Y. Hoshi, H. Miki, T. Takagi, *Diamond and Related Materials*, 17 (2008) 1669-1673.