Preventing Spin Relaxation of Optically Pumped Alkali Metal Atoms in Magnetometer by Atomically Thin Film Coating

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Abstract: We developed molecular layer deposition method of atomically thin hybrid polymer film for the first time by developing atomic layer deposition method with sequential surface chemical reactions in order to minimize the effect of the dipole-dipole interaction between the electron spin of alkali metal atoms and the nuclear spin of the atoms in the glass of the cell. We controlled film thickness of polymer thin film precisely and finally aimed at improving the sensitivity of the optically pumped atomic magnetometer. In the presentation, we report on the relaxation time of spin polarization by atomically thin hybrid polymer film with laser pump-probe method.

1 INTRODUCTION

The magneto-cardiogram test, which measures a very small magnetic field generated from the human heart with a highly sensitive magnetic sensor and performs two-dimensional mapping analysis, is expected to be able to evaluate cardiac electrical activity with higher spatial resolution and higher sensitivity than the electrocardiogram method in principle. On the other hand, extremely weak magnetic field measurement of fT order generated from neurons of the cerebral cortex is expected to play a very important role in noninvasively investigating human cranial nerve activity. At present, a superconducting quantum interference device (SQUID) having a sensitivity of 1 fT / $Hz^{1/2}$ order is used as a magnetic sensor for magneto-cardiography measurement and magneto-encephalography measurement. Using SQUID makes it possible to acquire knowledge about magnetic field mapping from the heart and brain and basic electrical activity in vivo, but since SQUID uses superconducting quantum interference effect, liquid helium It is necessary to operate it in an extremely low temperature state, and there is a problem that a large-sized apparatus becomes expensive maintenance cost. In recent years, attention has been paid to an optical pumping atomic magnetometer which measures a magnetic field by using spin polarization of alkali-metal atoms through the optical pumping method. The optical pumping method is a method in which light is used to create a large difference in the occupation number of atoms at two close energy levels and the optically pumped alkali metal atoms are spin polarized and the magnetic field applied there is linear. In order to rotate the plane of polarization of polarized light, we can estimate the magnetic field at room temperature from this angle of rotation. It has been energetically studied in the United States, Europe and the like, and at the National Institute of Standards and Technology (NIST), it has become compact to the size of a chip scale small watch (Schwindt et al., 2004).

In the optically pumped atomic magnetometer, the rotation angle of the polarization plane of the probe light becomes more sensitive to the change of the magnetic field as the relaxation rate of the spin becomes smaller. Recent reports from Princeton University reported that sensor sensitivity can reach sub fT / $Hz^{1/2}$ order by using SERF (Spin - Exchange - Relaxation - Free) state where the relaxation rate of spin polarization decreases (Kominis et al., 2003). Optical pumping atomic magnet sensors operating in the SERF state are expected. Therefore, it is extremely important to suppress the relaxation rate of spin polarization to a small value when spin-polarized atoms are used in an alkali vapor cell in order to measure with high precision. Although polarized spin is relaxed by collision of atoms against the inner wall

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of the cell, relaxation of spin polarization due to collision of this atom with the inner wall of the cell can be suppressed by coating the inner wall of the cell. This coating is called a spin relaxation preventing coating, and paraffin (CH_3 (C_nH_{2n}) CH_3: n> 20) has been widely used so far. The effect of prevention of spin relaxation by paraffin coating was first demonstrated by Robinson et al. in 1958 (Robinson et al., 1958). With no relaxation of spin polarization maximum of about 10,000, it is possible to collide with the inner wall, and it is expected to be applied to the field of ultrahigh sensitivity magnetic sensors and quantum communication. The well-known paraffin coating on the inner wall surface of the cell can be said to be an extremely useful means for obtaining a long spin relaxation time, but the physical action of its spin relaxation prevention effect is not well understood at present. According to Bouchiat et al. (Bouchiat et al., 1966), dipole-dipole interaction between electron spin of spin-polarized atom and nuclear spin of coated surface hydrogen atom, or orbital angular momentum and electron spin of relative motion between coating surface atoms and spin-polarized atoms Interaction is said to be the cause of spin relaxation. However, the result of this research alone is insufficient to understand the workings of the coating. For example, an effect of preventing spin relaxation at a high temperature is dulled, annealing at 80 °C for several hours in the presence of alkaline vapor called "aging process" after coating of paraffin increases spin relaxation prevention effect (Seltzer et al., 2010), but neither has been fully understood. Spin relaxation prevention effect obtained by the same coating material and the same manufacturing method is greatly different, and it falls within the skill of coating applicants and researchers.

One of authors has independently developed an atomic layer deposition method and an atomic layer deposition method of oxide by sequential surface chemical reaction using organometallic gas and water vapor as a starting material, so that an alkyl group (n = 1, 2, 3). We found that precise film thickness control can be realized using "self-limiting mechanism" appearing in the adsorption process, and in particular, we have found that it is possible to realize precise film thickness control using "self-limiting mechanism", to overcome the extremely difficult task of making a multilayer film structure (Kumagai et al., 1997).

2 EXPERIMENTAL SETUP

The experimental setup was comprised of a stainless steel vacuum chamber with two computer-controlled leak valves, a capacitor manometer, turbo-molecular pump (TMP) and quartz crystal unit to allow in situ measurements during growth of metal oxides. As a substrate, (100)-oriented Si wafers were used together with quartz glass cells. The substrate was first ultrasonically cleaned in conventional organic solvents, then dipped in 4.7% HF to remove the native oxide. After rinsing it in overflowing deionized water, it was loaded into a vacuum chamber. As vapor sources for the aluminum oxide film, two precursors were used in Fig.1. high-purity trimethyl-aluminum (TMA) and ethanol (EtOH) were used as precursor A and B, respectively.

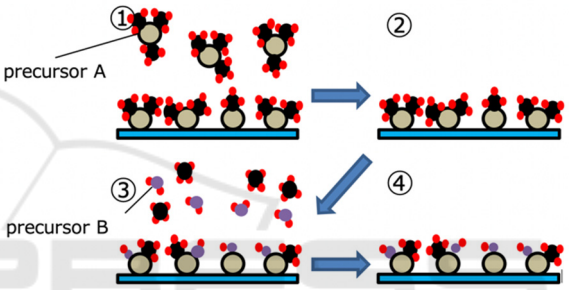

Figure 1: Atomic layer deposition utilizing two distinct precursors (A, B) sequentially dosed to the substrate producing a chemical reaction.

EtOH was prepared by Ethanol JIS special grade, ≥99.5%.These vapors were introduced alternately by two computer-controlled leak valves into the chamber which was evacuated by a TMP to a pressure below 10^{-7} Torr. Figure 2 shows input signals applied to the computer-controlled leak valves for generating each vapor pulse whose peak vapor pressure reached $1x10^{-4}$ Torr. The duration of supplying each vapor pulse was 30 s, while the chamber was continuously exhausted during the growth. The time point exactly 20 s before the first dosing of TMA vapor was defined as t= 0 s. Therefore, TMA vapor was first introduced at t = 20 s during the supplying time, and then at t= 110 s, EtOH vapor was first introduced, and then at t= 200 s, TMA vapor was introduced again. From t = 290 s onwards, these binary vapors were supplied alternately according to the sequence in Fig. 2.

Atomic layer deposition at room temperature was carried out by changing the combination of trimethylaluminum (TMA) and water vapor (H_2O) which was often used in the atomic layer deposition method, in addition to H_2O as ethanol as an oxidizing

agent. Assuming that the introduction cycle period is 6 s, TMA was introduced to the first 0 - 1 s and an oxidizing agent was introduced to 3 - 4 s. The peak pressure of the material gas to be introduced was kept constant at 10^{-4} Torr, the total time of introduction of the source gas was kept constant, the introduction pulse time and the number of cycles were changed to seven types, and the deposition characteristics were investigated. All atomic layer deposition processes were done at room temperature and a thin film was deposited on the substrate. The film thickness and the refractive index were measured with a spectroscopic ellipsometer.

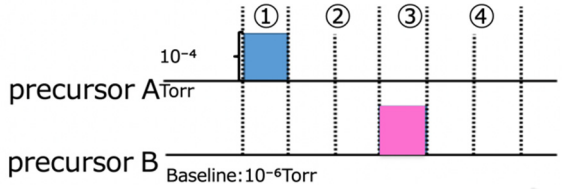

Figure 2: Input signals applied on the computer-controlled leak valves for generating each vapor pulse.

3 MOLECULAR LAYER DEPOSITION OF ORGANIC-INORGANIC HYBRID POLYMER THIN FILM REALIZED

Figure 3 shows variations of vapor pressure in the vacuum chamber and film thickness with growth time at room temperature. The vapor pressures in the vacuum chamber evacuated by a TMP to a pressure below 10^{-7} Torr follows the input signals in Fig. 2. Duration of each pulse of vapor pressure was 30 s. The film thickness also shown in Fig. 3 indicates the thickness from the initial surface (t = 0 s). The duration of supply of TMA and EtOH vapors which were introduced in the sequence shown in Fig. 3, was 30 s. Figure 3 shows the case when binary vapors of TMA and EtOH were supplied by taking into account the sequence shown in Fig. 2, an increase of around 1 nm is found to occur upon the introduction of TMA. This is because dosing of TMA to surface -O-H groups causes chemical reactions by which OH groups change into -O-Al-CH_3 groups, whereas dosing of EtOH to surface Al- CH_3, groups causes chemical reactions in which Al- CH_3 groups change into Al-O-H groups. Although actual surface reactions must be more complicated than the simplified picture mentioned here, the picture can also be supported by infrared spectroscopic studies.

Modification of O-H-terminated surfaces to -O-Al-CH_3 makes the increase of thickness 0.1 nm greater than modification of Al-CH_3-terminated surfaces to Al-O-H. It was found in Fig. 3 that the film thickness slightly decreased just after the increase, because high vacuum caused desorption of molecules which had adsorbed at room temperature. Growth rates were 0.887 nm/cycle, the same as obtained by dividing the total thickness of the film from an ex situ variable-angle spectroscopic ellipsometer (LA. Woollam Co., Inc.) by the number of growth cycles. This exhibits characteristics of self-limiting nature of adsorption which are characteristic of the growth technology in this study.

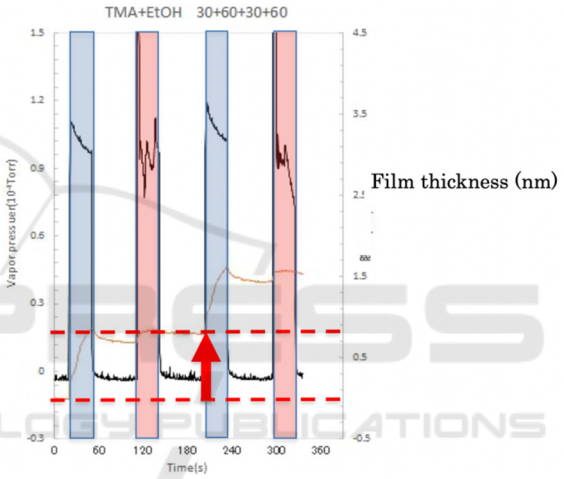

Figure 3: Variations of vapor pressure in the vacuum chamber and film thickness with growth time at room temperature.

4 MEASURE SPIN POLARIZATION RELAXATION TIME

To characterize the quality of the film fabricated on inner-wall of the glass cell, we performed measurements of the relaxation time of the optical pumped rubidium atoms with the pump probe method in Fig.4. The laser frequency of the pump and probe light beams was same and then tuned to be resonant to $5S_{1/2}$ (F = 3) -> $5P_{3/2}$ (F' = 2, F' = 3, F' = 4) of ^{85}Rb optical transition, until a maximum of fluorescent intensity in the separate, uncoated cell was obtained. Then, the pump beam in the cell was supplied by abrupt opening of a shutter. The atoms on the Rb atom ground state $5S_{1/2}$ (F = 3) were excited by the

radiation; and the radiation populated $5S_{1/2}$ (F = 2) ground state through the intermediate atomic upper states $5P_{3/2}$ (F' = 1, F' = 2, F' = 3, F' = 4). As a result of the optical pumping process, the amplitude of $5S_{1/2}$ (F = 3) line decreased while the amplitude of $5S_{1/2}$ (F = 2) line increased.

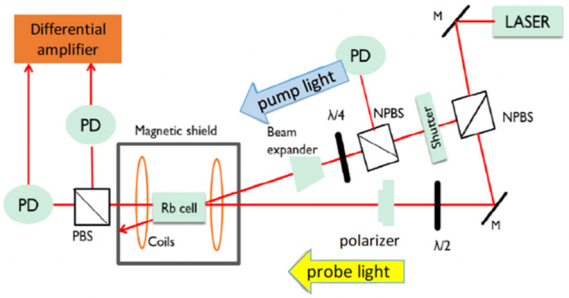

Figure 4: Schematic of the setup for spin relaxation measurement.

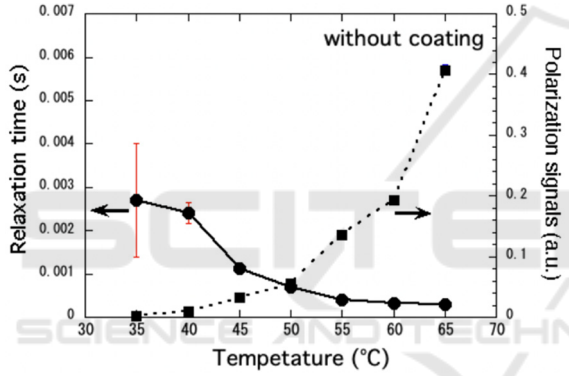

Figure 5: Temperature dependencies of spin relaxation time and polarization signal in a Rb cell without coating.

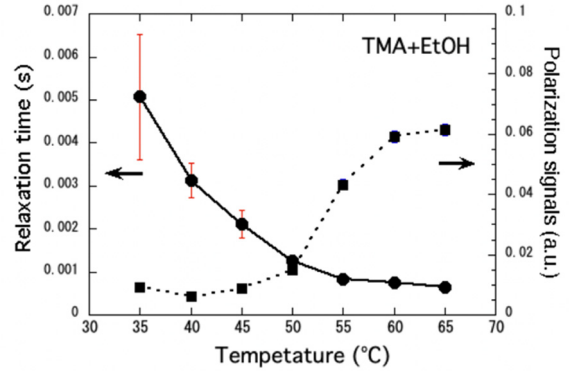

Figure 6: Temperature dependencies of spin relaxation time and polarization signal in a Rb cell with thin film coated by sequentially dosing TMA and ethanol vapor.

Figure 5 shows temperature dependencies of spin relaxation time and polarization signal in a Rb glass cell without coating. At 35°C, the relaxation time was estimated as 2.7 ms. In a Rb cell with thin film coated by sequentially dosing TMA and ethanol vapor, temperature dependencies of spin relaxation time and polarization signal are shown in Fig. 6.

The relaxation time in the glass cell with coating was higher than those without coating. At 35°C, it was more than 5 ms, twice higher than that without coating.

5 CONCLUSIONS

We controlled film thickness of hybrid polymer thin film precisely by developing atomic layer deposition method with sequential surface chemical reactions and then could improve the relaxation time of spin polarization by coating with the thin film. Although the physical action of its spin relaxation prevention effect is not well understood at present, it will definitely improve the sensitivity of the optically pumped atomic magnetometer.

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