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Title	Co thickness dependence of structural and magnetic properties in spin quantum cross devices utilizing stray magnetic fields
Author(s)	Kaiju, Hideo; Kasa, Haruya; Komine, Takashi et al.
Citation	Journal of applied physics, 117(17), 17C738-1-17C738-4 https://doi.org/10.1063/1.4917061
Issue Date	2015-05-08
Doc URL	https://hdl.handle.net/2115/59519
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Type	journal article
File Information	1.4917061.pdf

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AIP Journal of Applied Physics

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Citation: Journal of Applied Physics **117**, 17C738 (2015); doi: 10.1063/1.4917061
View online: http://dx.doi.org/10.1063/1.4917061
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JOURNAL OF APPLIED PHYSICS **117**, 17C738 (2015)

Co thickness dependence of structural and magnetic properties in spin quantum cross devices utilizing stray magnetic fields

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(Presented 7 November 2014; received 18 September 2014; accepted 3 December 2014; published online 7 April 2015)

We investigate the Co thickness dependence of the structural and magnetic properties of Co thin-film electrodes sandwiched between borate glasses in spin quantum cross (SQC) devices that utilize stray magnetic fields. We also calculate the Co thickness dependence of the stray field between the two edges of Co thin-film electrodes in SQC devices using micromagnetic simulation. The surface roughness of Co thin films with a thickness of less than 20 nm on borate glasses is shown to be as small as 0.18 nm, at the same scanning scale as the Co film thickness, and the squareness of the hysteresis loop is shown to be as large as 0.96–1.0. As a result of the establishment of polishing techniques for Co thin-film electrodes sandwiched between borate glasses, we successfully demonstrate the formation of smooth Co edges and the generation of stray magnetic fields from Co edges. Theoretical calculation reveals that a strong stray field beyond 6 kOe is generated when the Co thickness is greater than 10 nm at a junction gap distance of 5 nm. From these experimental and calculation results, it can be concluded that SQC devices with a Co thickness of 10–20 nm can be expected to function as spin-filter devices. © *2015 AIP Publishing LLC*.
[http://dx.doi.org/10.1063/1.4917061]

I. INTRODUCTION

Spintronics is an emerging research field which utilizes the charge and spin degrees of freedom of electrons in solid-state systems.[1–4] One of the fascinating phenomena in spintronics is the spin-filter effect, which allows us to obtain highly spin-polarized charge carriers in nonmagnetic materials.[5–9] Recently, we proposed spin quantum cross (SQC) devices utilizing stray magnetic fields as a new type of spin-filter device.[10] This SQC device consists of inorganic complexes or quantum dots (QDs) sandwiched between two crossed edges of magnetic thin-film electrodes. In this structure, a high magnetic field could be locally generated in the inorganic complexes or QDs due to the contributions of the stray field from both edges of the magnetic thin-film electrodes. Because a large magnetic field produces a large Zeeman effect, energy splitting of the inorganic complexes or QDs can be enhanced. Therefore, a large spin-filter effect can be expected. In our previous work, we reported some preliminary results for SQC devices. Typical results included the observation of ohmic characteristics in Ni/Ni devices, nanoscale tunneling phenomena in Ni/NiO/Ni devices, and the ballistic regime of nanoscale molecules in Ni/P3HT:PCBM/Ni devices.[11–13] Although such nanoscale phenomena have been observed in SQC devices, these devices cannot function as spin-filter devices, because the squareness of the hysteresis loop of magnetic thin films on polyethylene naphthalate (PEN) substrates, used in the magnetic electrodes of SQC devices, is not sufficiently large.[14] In contrast, our ongoing research results suggest that Co thin films on borate glasses are good candidates for magnetic electrodes in SQC devices. However, the detailed structural and magnetic properties of such electrodes have not been clarified, especially in terms of their dependences on the Co film thickness. In this study, we investigate the dependences of the structural and magnetic properties of Co thin films on its thickness on borate glasses and Co thin-film electrodes sandwiched between borate glasses in SQC devices, and also calculate the dependence of the stray field between the two edges of Co thin-film electrodes on the Co film thickness in SQC devices using micromagnetic simulation.

II. EXPERIMENTAL

Co thin films were deposited on borate-glass substrates with a glass transition temperature T_g of 464 °C (ISUZU GLASS CO., LTD.) under a magnetic field of 400 Oe by electron beam evaporation. The growth rate of the Co thin films was 0.15 nm/s at an evaporation power of 160–200 W. The dimensions of the borate-glass substrates were $10 \times 10 \times 2\,\mathrm{mm}^3$, and both sides were obliquely cut and polished before the deposition of the Co thin films. The Co thin films deposited on the borate-glass substrates were stacked on borate glasses of the same composition using a glass mold pressing technique at a glass deformation temperature A_t of 503 °C under a pressure of 0.25 MPa and then cut down to half of the samples. Finally, the cross-sectional surfaces of the stacked samples were polished by a mechanical polishing (MP) method using Al_2O_3-based emeries and a chemical mechanical polishing (CMP) method using CeO_2 and Al_2O_3 slurries. The surface morphologies and roughnesses of the samples were analyzed by atomic force microscopy (AFM;

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Nanonavi IIs, SII NanoTechnology). Microstructures were evaluated using scanning electron microscopy (SEM; JSM-6700FT, JEOL) operating at 5 kV. The magnetization curves were measured using the focused magneto-optical Kerr effect (MOKE; BH-PI920-HU, NEOARK) under a magnetic field of up to 1 kOe at room temperature. The stray fields originating from the edges of the Co thin films were observed by magnetic force microscopy (MFM), in which a CoCrPt-coated cantilever was used in the AFM setup described above. The local magnetic fields generated between the two edges of the Co thin-film electrodes in SQC devices were calculated by micromagnetic simulation.

III. RESULTS AND DISCUSSION

Figure 1(a) shows the surface roughness as a function of the Co thin film thickness on borate glasses. For comparison, the surface roughness of Co thin films on PEN substrates, which have also been examined as magnetic thin-film electrodes of SQC devices,[14] is also shown. The insets show AFM surface images of Co thin films on borate glasses and the borate glass without a Co thin film. The scanning area is $500 \times 500\,\mathrm{nm}^2$. Here, the surface roughness R_a is defined by $R_a = 1/L_xL_y \int_0^{L_x}\int_0^{L_y} |h(x,y)|dxdy$, where h (x,y) is the height profile as a function of x and y, and $L_{x(y)}$ is the lateral scanning size along the x (y) direction. The surface roughness of Co thin films on borate glasses is as small as 0.44–0.65 nm. From Fig. 1(a), it is found that this surface roughness value is smaller than that of Co thin films on PEN substrates, 0.79–1.3 nm. The realization of smooth Co thin films is attributed to the use of borate glass substrates with a smooth surface and a low growth rate of Co thin films. The vaporized Co atoms can move easily and freely along the in-plane direction on the borate glass substrate with a smooth surface. Under this situation, the Co atoms can diffuse on the surface for a sufficiently long time owing to the low growth rate of 0.15 nm/s. This makes it possible to form smooth surface structures, which are favored by surface energy minimization. Figure 1(b) shows the scaling properties of the surface roughness of Co thin films on borate glasses and the borate glass without the Co film. Because the junction area in SQC devices is determined by the film's thickness, the surfaces need to be smooth enough at the same scanning scale as the film thickness. As shown in Fig. 1(b), the surface roughnesses of a Co thin film with a thickness of 16 nm on a borate glass and that of the borate glass without the Co film are as small as 0.15 nm and 0.08 nm, respectively, corresponding to less than a few atomic layers, at a scanning scale of 16 nm. The surface roughness of a Co thin film with a thickness of less than 21 nm on a borate glass is less than 0.18 nm at the same scanning scale as the Co film thickness. These results indicate that Co thin films on borate glasses are good candidates for magnetic thin-film electrodes in SQC devices from the perspective of surface structures.

Figure 2(a) shows the magnetization curves of Co thin films on borate glasses obtained by MOKE measurements. The Co thicknesses are 8.5, 11, 15.9, and 17.5 nm, respectively. The magnetic field is applied in the magnetic easy-

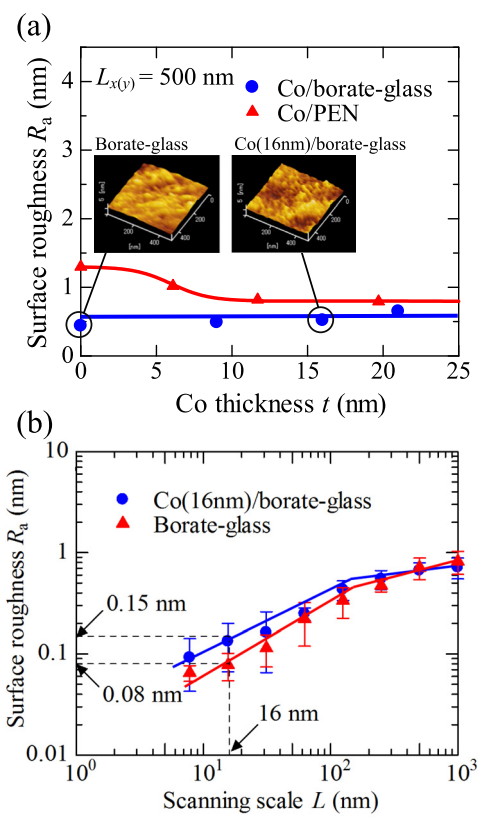

FIG. 1. (a) Surface roughness as a function of the Co thickness in Co/borate-glass and Co/PEN and (b) scaling properties of the surface roughness of Co(16 nm)/borate-glass and borate-glass without the Co film.

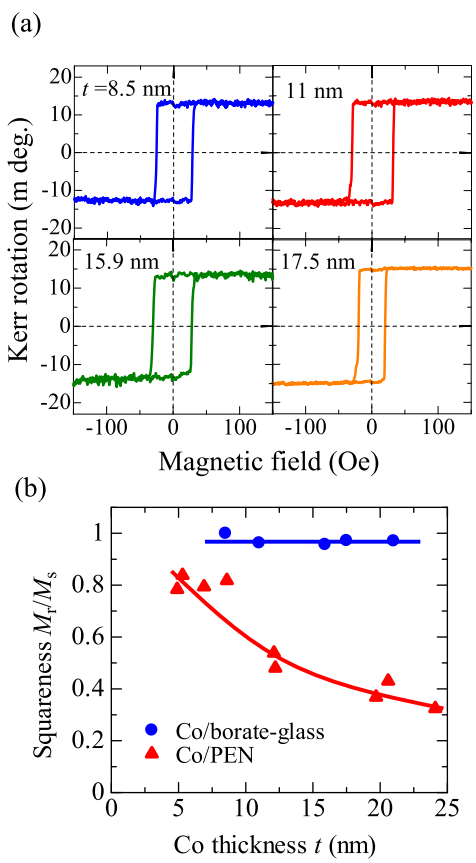

FIG. 2. (a) Magnetization curves of Co/borate-glass and (b) M_r/M_s as a function of the Co thickness in Co/borate-glass and Co/PEN.

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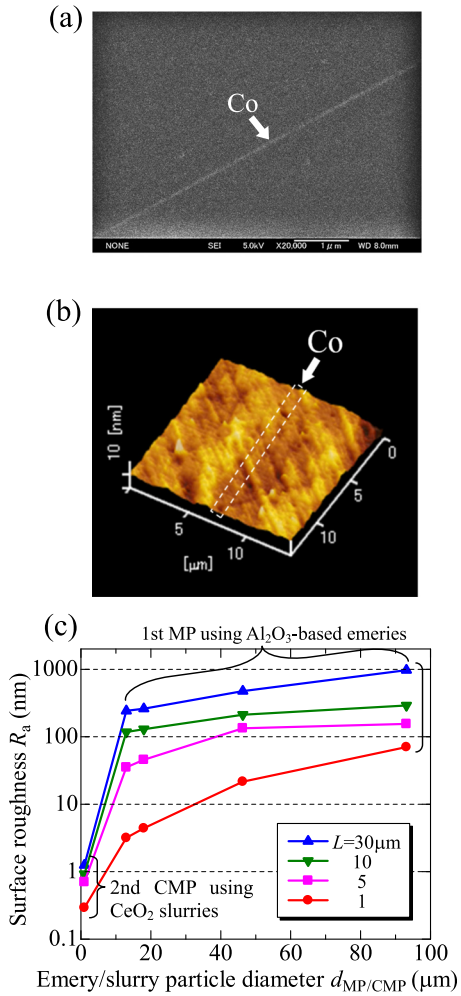

FIG. 3. (a) SEM image and (b) AFM image of the polished surfaces of borate-glass/Co/borate-glass. (c) Surface roughness of the polished surfaces of borate-glass/Co/borate-glass as a function of the particle diameter $d_{MP/CMP}$ of Al_2O_3-based emeries ($d_{MP} > 10\,\mu m$) or the CeO_2 slurries ($d_{CMP} = 1\,\mu m$).

axis direction. The coercive force of a Co thin film on a borate glass is 20–30 Oe, which is almost the same as the value reported for Co films.[15] The squareness of the hysteresis loop M_r/M_s, where M_r and M_s are the remanent and saturation magnetizations, respectively, is as large as 0.96–1.0. The squareness as a function of the Co thickness is shown in Fig. 2(b). The squareness of the hysteresis loop of Co thin films on borate glasses is greater than that of Co thin films on PEN substrates, 0.32–0.84. This large squareness causes a high stray field to be generated from the edges of Co thin-film electrodes in SQC devices. These results indicate that Co thin films on borate glasses are suitable for use as magnetic thin-film electrodes in SQC devices from the perspective of not only the surface structures but also the magnetic properties.

Figure 3 shows an SEM image of the polished surfaces of Co thin-film electrodes sandwiched between the two borate glasses. The thickness of the Co thin films is 14 nm. From this image, Co edges can be clearly confirmed. The contrast enhancement of the Co edges is attributed to the high conductivity of Co. There is no structural difference around the Co edges, which is confirmed by the AFM observation, as shown in Fig. 3(b). As can be seen from Fig. 3(b), the Co edges are not recognized, although the Co edges exist at the center of the AFM image. This means that there is no step difference between Co thin-film electrodes and borate glasses. The roughness of the polished surfaces of Co thin-film electrodes sandwiched between borate glasses is as small as 0.29–1.2 nm in a scanning area of 1×1–$30 \times 30\,\mu m^2$. The high conductivity and small surface roughness of the Co edges are realized by a three-step polishing process using MP and CMP methods, which are described below. Figure 3(c) shows the roughness of the polished surfaces of Co thin-film electrodes sandwiched between borate glasses as a function of the particle diameters d_{MP} (the MP process) and d_{CMP} (the CMP process). The d_{MP} of the Al_2O_3-based emeries in the first MP process is greater than 10 μm, and d_{CMP} of CeO_2 slurries in the second CMP process is equal to 1 μm. The first MP process is performed using Al_2O_3-based emeries in the following sequence of particle diameters: 93, 46, 18, and 13 μm. Thereafter, the second CMP process is performed using CeO_2 slurries with a particle diameter of 1 μm. From Fig. 3(c), it is found that the surface roughness decreases when the particle diameter of the emeries/slurries decreases and is as small as 0.29–1.2 nm in the second CMP process, where CeO_2 slurries with a particle diameter of 1 μm are used. The third CMP process is performed using Al_2O_3 slurries with a particle diameter of 0.1 μm. This third process causes a high increase in the conductivity of the Co edges. These established techniques for the MP and CMP methods enable the generation of a stray field from the Co edges and its observation by MFM. Figure 4 shows an MFM image of the polished surfaces of Co thin-film electrodes sandwiched between borate glasses. The thicknesses of the Co thin films are 8.5, 11, and 15 nm, respectively. Blue colored bars in the MFM images indicate that the stray field is generated along the out-of-plane direction of the samples. As can be seen from Fig. 4, a strong

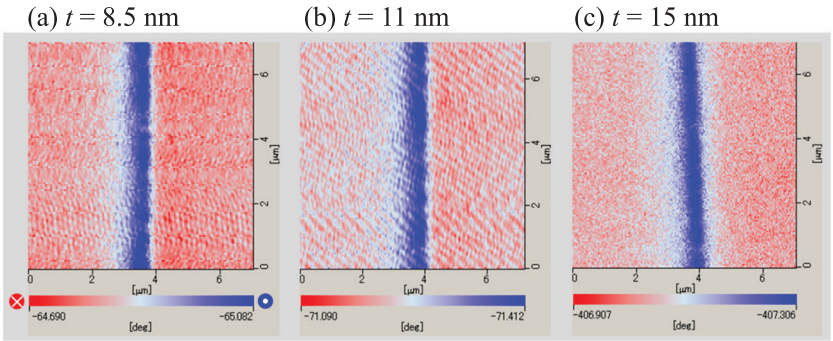

FIG. 4. MFM images of the polished surfaces of borate-glass/Co/borate-glass; Co thicknesses are (a) 8.5, (b) 11, and (c) 15 nm, respectively.

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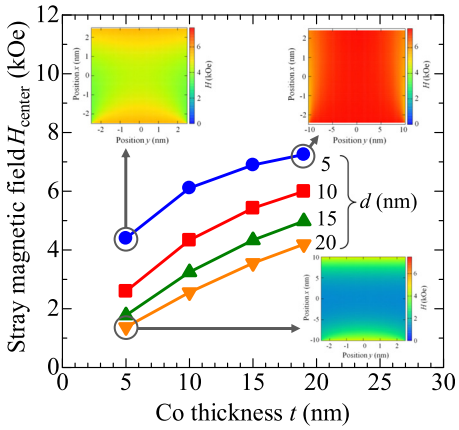

FIG. 5. Calculation results of the Co thickness dependence of the stray magnetic field between the two edges of Co thin-film electrodes in SQC devices. The insets show the distribution of the stray field; $t = 5$ nm and $d = 20$ nm, $t = 5$ nm and $d = 5$ nm, and $t = 20$ nm and $d = 5$ nm.

stray field is generated along the out-of-plane direction from the Co edges. This behavior is consistent with the results obtained by MOKE measurements, indicating that the squareness of the hysteresis loop is as large as 0.96–1.0, as shown in Fig. 2. Here, we note that that the width of the stray field is about 0.5–1 μm, which is much larger than the Co thicknesses of 8.5, 11, and 18 nm. This is attributed to a long spacing distance (~50 nm) between the CoCrPt-coated cantilever of the MFM setup and the edge of the Co thin-film electrode, which prevents the magnetization reversal of the CoCrPt-coated cantilever. Thus, we have successfully demonstrated that Co thin-film electrodes sandwiched between borate glasses with a flat edge surface and a strong stray field generating from the edges can be fabricated using our proposed technique.

Figure 5 shows the results of calculations of the Co thickness dependence of the stray magnetic field between the two edges of the Co thin-film electrodes in SQC devices. The insets show the distribution of the stray field for varying Co thicknesses t and distances between the two edges of the Co thin-film electrodes d; specifically, $t = 5$ nm, $d = 20$ nm; $t = 5$ nm, $d = 5$ nm; and $t = 20$ nm, $d = 5$ nm. The points x and y are the positions along the in-plane and out-of-plane directions of the Co films, respectively, between the two edges of the Co thin-film electrodes. The position $x = y = 0$ nm is the center of the junction between the two edges of the Co thin-film electrodes. At $t = 5$ nm and $d = 20$ nm, the stray field is less than 2 kOe, and a uniform distribution is not obtained. At $t = 5$ nm and $d = 5$ nm, although the stray field is beyond 4 kOe, a uniform distribution is still not obtained. At $t = 20$ nm and $d = 5$ nm, the stray field measures as high as 7 kOe, and a uniform distribution is successfully obtained. From the calculation plots of Fig. 5, it is found that a high stray field beyond 6 kOe is generated when the Co thickness is larger than 10 nm and the distance between the two edges of the Co thin-film electrodes is 5 nm. This high magnetic field produces a large Zeeman effect in the sandwiched materials between the two edges of the magnetic thin-film electrodes. These experimental and calculation results conclude that SQC devices with Co thin-film electrodes sandwiched between borate glasses (Co thickness $t = 10$–20 nm) can be used as spin-filter devices.

ACKNOWLEDGMENTS

This research has been partially supported by the Precursory Research for Embryonic Science and Technology program from the Japan Science and Technology Agency (JST), a Grant-in-Aid for Young Scientists from the Japan Society for the Promotion of Science (JSPS), and the Cooperative Research Program of the Network Joint Research Center for Materials and Devices from the Ministry of Education, Culture, Sports, Science and Technology (MEXT). The authors would like to express their sincere appreciation to T. Ohta and M. Takei of Hokkaido University, N. Yamashita of ISUZU GLASS CO., LTD., and T. Yamaoka of Hitachi High-Tech Science Corporation for their cooperation and helpful discussions.

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