The dependence of nano-contact magnetoresistance on the bulk scattering spin asymmetry in CoFe alloys with oxidation impurities

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Nano-contact magnetoresistance (NCMR) spin-valves (SVs) using an AlO_x nano-oxide-layer (NOL) have numerous nanocontacts in the thin AlO_x oxide layer. The NCMR theoretically depends on the bulk scattering spin asymmetry (β) of the ferromagnetic material in the nanocontacts. To determine the relationship between NCMR and β , we investigated the dependence of NCMR on the composition of the ferromagnetic material $\text{Co}_{1-x}\text{Fe}_x$. The samples were annealed at 270 °C and 380 °C to enhance the MR ratio. For both annealing temperatures, the magnetoresistance ratio in the low-resistance area product region at less than $1 \Omega \mu\text{m}^2$ was maximized for $\text{Co}_{0.5}\text{Fe}_{0.5}$. To evaluate β exactly, we fabricated current-perpendicular-to-plane giant magnetoresistance SVs with $\text{Co}_{1-x}\text{Fe}_x/\text{Cu}/\text{Co}_{1-x}\text{Fe}_x$ layers and used Valet and Fert's theory to solve the diffusion equation of the spin accumulation for a ferromagnetic layer/non-ferromagnetic layer of five layers with a finite diffusion length. The evaluated β for $\text{Co}_{1-x}\text{Fe}_x$ was also maximized for $\text{Co}_{0.5}\text{Fe}_{0.5}$. Additionally, to determine the difference between the experimental MR ratio of NCMR SVs and the theoretical MR ratio, we fabricated $\text{Co}_{0.5}\text{Fe}_{0.5}$ with oxygen impurities and estimated the decrease in β with increasing oxygen impurity concentration. Our $\text{Co}_{0.5}\text{Fe}_{0.5}$ nanocontacts fabricated using ion-assisted oxidation may contain oxygen impurities, and the oxygen impurities might cause a decrease in β and the MR ratio. © 2015 AIP Publishing LLC.
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I. INTRODUCTION

Nano-contact magnetoresistance (NCMR) has attracted much interest for its potential applications in spin-electronic devices such as spin torque oscillators^{1,2} and magnetic sensors. NCMR spin-valves (SVs) are fabricated using an aluminum oxide (AlO_x) nano-oxide layer (NOL) that serves as a spacer layer for the current-perpendicular-to-plane (CPP) structures. Such a NOL has many ferromagnetic conductive channels called NCs. For a thin AlO_x plane, these channels are roughly 2 nm in diameter.³ Recently, NCMR in a $\text{Fe}_{0.5}\text{Co}_{0.5}\text{-AlO}_x$ NOL was experimentally shown and an MR ratio of 35% at a resistance area product (RA) of $0.3 \Omega \mu\text{m}^2$ was reported.⁴

The origin of the NCMR was attributed to spin accumulation and mistracking effects⁵ around nanometer-sized domain walls in the ferromagnetic nano-contact in the anti-parallel magnetization configuration. This occurs between the free and pinned layers.⁶ In Ref. 5, the difference in the conductivity between spin-up (+) and spin-down (−) because of mistracking around the domain wall in the NC was shown using the following equation:

$$\sigma^\pm = \sigma^{\pm(0)} \left[1 \mp 2\zeta^2 \left(\frac{3\beta}{5(1 \mp \beta)} - \frac{2\beta}{3\sqrt{1 - \beta^2}} \right) \right], \quad (1)$$

where $\sigma^{\pm(0)}$ represents the spin-up (+) and spin-down (−) conductivity of the bulk material, $\zeta = \pi\hbar^2 k_F / 4mhJ$ is the perturbation coefficient, $\hbar$ is Planck's constant, k_F is the Fermi wave number, m is the electron mass, J is the

exchange coupling constant, and h is domain wall length. β is the bulk scattering spin asymmetry coefficient of the ferromagnet. Simulated results showed that the MR ratio increased because of the mistracking effect of the electrons caused by domain walls in the NCs with increasing β . That is, the NCMR depends strongly on the value of β for the ferromagnet in the NC. However, it is well known that tunneling magnetoresistance (TMR) is generally related to the spin polarization of ferromagnetic materials according to Julliere's model.⁷ Previous investigations reported the relationship between TMR and spin polarization of the ferromagnet experimentally in $\text{Co}_{1-x}\text{Fe}_x$.⁸ In NCMR, using $\text{Co}_{1-x}\text{Fe}_x$ for the ferromagnetic NCs may be used to determine the relation between NCMR and $\text{Co}_{1-x}\text{Fe}_x$ and may also identify the relationship between NCMR and β of the ferromagnets in NCs.

In a previous study, the β values of some CoFe alloys were evaluated using CPP giant magnetoresistance (GMR) methods using Valet-Fert's (VF) theory.⁹ However, there have been very few studies on how the Fe composition of the $\text{Co}_{1-x}\text{Fe}_x$ alloy relates to β . Additionally, the β values obtained for each group using VF theory differed despite using the same ferromagnetic material, for example, $\text{Co}_{0.9}\text{Fe}_{0.1}$.^{10,11} The most likely reason for this is that different parameters in the VF model were used for evaluating the value of β . To evaluate the value of β , Reilly *et al.* reported the influence of the spin-diffusion length (ℓ_{sf}) on ΔRA for a thick ferromagnetic layer. The exact fitting curves of ΔRA as a function of the thickness of the ferromagnet were obtained by using the finite spin-diffusion length.¹¹ Additionally,

Taniguchi *et al.* considered not only the finite spin-diffusion length but also solved the diffusion equation for spin accumulation without assuming a periodic multilayer.¹² Therefore, some ferromagnetic materials cannot be compared using their β values without a common experimental method and analysis based on the VF model.

We have already reported the existence of oxidation impurities in $\text{Fe}_{0.5}\text{Co}_{0.5}$ NCs from X-ray Photoelectron Spectroscopy (XPS) analysis and the resistivity of the NCs.³ It is possible that oxygen impurities can cause spin-independent scattering as indicated by the increase in the resistivity of the material. That is, oxidation impurities can decrease the β value of FeCo.

In this paper, we first show investigations of the dependence of NCMR of $\text{Co}_{1-x}\text{Fe}_x$ alloys with $x=0.1-0.7$. We then evaluated the β values of a CoFe alloy to determine a model that most accurately solves the diffusion equation of spin accumulation in five Ferro-/Non-Ferromagnetic (F/N) layers with a finite diffusion length based on VF theory. Finally, we considered the effect of oxygen impurities on the β value in a $\text{Fe}_{0.5}\text{Co}_{0.5}$ alloy.

II. EXPERIMENTAL

Films were prepared using ion-beam sputtering and DC magnetron sputtering. We prepared three types of spin valves (Fig. 1): NCMR SVs and CPP-GMR SVs for evaluating β with the $\text{Co}_{1-x}\text{Fe}_x$ alloy and CPP-GMR SVs to estimate the effect of oxidation impurities. The NCMR top SV had the structure SiO_x substrate/Cu electrode/Ru (2 nm)/ $\text{Co}_{1-x}\text{Fe}_x$ t^{NCMR} /Al (1.3 nm)/oxidation process/ $\text{Co}_{1-x}\text{Fe}_x$ t^{NCMR} /IrMn (7 nm)/capping layer. The $\text{Co}_{1-x}\text{Fe}_x$ alloys used were $\text{Co}_{0.9}\text{Fe}_{0.1}$, $\text{Co}_{0.7}\text{Fe}_{0.3}$, $\text{Co}_{0.5}\text{Fe}_{0.5}$, and $\text{Co}_{0.3}\text{Fe}_{0.7}$. The compositions of the films were determined to be $\text{Co}_{0.86}\text{Fe}_{0.14}$, $\text{Co}_{0.67}\text{Fe}_{0.33}$, $\text{Co}_{0.49}\text{Fe}_{0.51}$, and $\text{Co}_{0.71}\text{Fe}_{0.29}$, respectively. To ensure that the product of the magnetization and the ferromagnetic thickness (t^{NCMR}) remains constant, thicknesses of 3.0, 2.6, 2.4, and 2.4 nm were used for the $\text{Co}_{1-x}\text{Fe}_x$ alloy. NCs in

the AlO_x NOL were formed using ion-assisted-oxidation (IAO), which involves oxygen exposure with an Ar^+ ion beam irradiated on the Al surface. The intensity of the IAO was controlled using the irradiation time under a constant O_2 flow of 1.0 sccm. After deposition, SVs were annealed in a field of 10 kOe, first at 270 °C for 1.5 h and then at 380 °C for 1.5 h. We measured the MR ratio and the RA of NCMR using the current in plane tunneling (CIPT) method.

The CPP-GMR bottom SV had the structure SiO_x substrate/Cu electrode/seed layer/IrMn (7 nm)/ $\text{Co}_{1-x}\text{Fe}_x$ (3 nm)/Cu (5 nm)/ $\text{Co}_{1-x}\text{Fe}_x$ (t_{F1})/Ru (2 nm)/capping layer. The thickness of the pinned layer was fixed at 3 nm to maintain a high exchange field (H_{ex}) and the thickness of the free layer varied between $t_{\text{F1}} = 2$ and 20 nm. These samples were patterned into pillars using electron beam (EB)-lithography and Ar ion milling. CPP-GMR SVs were annealed in a field of 10 kOe at 270 °C for 1.5 h after deposition. The diameter of the CPP pillar was between 0.7 and 1.5 μm . The pillar sizes were observed using scanning-electron-microscopy (SEM). The CPP-GMR characteristics were measured using the DC four probe method at room temperature. β and the interface scattering spin asymmetry (γ) were determined by solving the diffusion equation for spin accumulation in a five-layer F/N system with a finite spin-diffusion length based on VF theory as described in Ref. 12. In this study, we re-calculated the five-layer VF model instead using an asymmetric model with free and pinned CoFe layers of unequal thickness. We fit the product of ΔR and the pillar size (ΔRA) as a function of the thickness of the free layer with this VF model to obtain β and γ . For an estimate of the resistivity of the CoFe alloy, we fabricated the CIP (current-in-plane) elements with the structure SiO_x substrate/Ta(3 nm)/Ru(2 nm)/CoFe alloy (6 nm, 8 nm, and 10 nm)/Ta(5 nm).

To determine the effect of oxygen impurities in $\text{Co}_{0.5}\text{Fe}_{0.5}$, we fabricated a CPP-GMR SV structure of SiO_x substrate/Cu electrode/seed layer/IrMn (7 nm)/ $\text{Co}_{0.5}\text{Fe}_{0.5}$ (3 nm)/Cu (5 nm)/ $\text{Co}_{0.5}\text{Fe}_{0.5}$ with oxygen impurity/Ru (2 nm)/capping layer. The $\text{Co}_{0.5}\text{Fe}_{0.5}$ layer with oxygen impurities

FIG. 1. Schematic of the devices: (a) NCMR SV; (b) CPP-GMR-SV used for evaluating β for the $\text{Co}_{1-x}\text{Fe}_x$ alloy; (c) CPP-GMR-SV used to obtain an estimate of the effect of the oxidation impurities.

was produced as a multi-layer of $\text{Co}_{0.5}\text{Fe}_{0.5}$ t_{F2} /Natural Oxidation (NO) (O_2 pressure: 2.92×10^{-3} Pa applied for 2 s) for n layers. We controlled the magnitude of the oxygen impurities in $\text{Co}_{0.5}\text{Fe}_{0.5}$ with the thickness of $\text{Co}_{0.5}\text{Fe}_{0.5}$ t_{F2} and fabricated the three types of $\text{Co}_{0.5}\text{Fe}_{0.5}$ with oxidation impurities, abbreviated as FeCoO1, FeCoO2, and FeCoO3. The specific details of each sample are as follows: FeCoO1: $\text{Co}_{0.5}\text{Fe}_{0.5}$ (2 nm)/NO; FeCoO2: $\text{Co}_{0.5}\text{Fe}_{0.5}$ (1.5 nm)/NO; FeCoO3: $\text{Co}_{0.5}\text{Fe}_{0.5}$ (1 nm)/NO. For an estimate of β from the five-layer VF model, we varied the total free thickness of $\text{Co}_{0.5}\text{Fe}_{0.5}$ with oxygen impurities as a function of the number of multilayers, n . These samples were annealed in a field of 10 kOe at 270°C for 1.5 h after deposition. CPP elements and CIP elements for an estimate of the resistivity were fabricated using the previously described method.

III. RESULTS AND DISCUSSION

We fabricated NCMR SVs with $\text{Co}_{1-x}\text{Fe}_x$ NCs with an AlO_x NOL. NCs were fabricated by the oxidation of metal Al on a CoFe alloy using the IAO method.³ After IAO, the same CoFe alloy was deposited on the NOL. Figure 2 shows the relationship between the MR ratio and RA for $\text{Co}_{0.9}\text{Fe}_{0.1}$, $\text{Co}_{0.7}\text{Fe}_{0.3}$, $\text{Co}_{0.5}\text{Fe}_{0.5}$, and $\text{Co}_{0.3}\text{Fe}_{0.7}$. In each CoFe alloy, various IAO times (30–120 s) required for oxidation were used to obtain various RA. We fit the MR ratio as a function of RA for samples annealed at 270°C using a parallel-circuit model¹³ using the following equations:

$$RA_{\min} = RA_{\text{para}} + \frac{RA_{\text{NOL}} \cdot RA_{\text{NC}}}{RA_{\text{NOL}} \cdot D + RA_{\text{NC}}(1 - D)}, \quad (2)$$

$$RA_{\max} = RA_{\text{para}} + \frac{RA_{\text{NOL}}(1 + MR_{\text{NOL}}) \cdot RA_{\text{NC}}(1 + MR_{\text{NC}})}{RA_{\text{NOL}}(1 + MR_{\text{NOL}}) \cdot D + RA_{\text{NC}}(1 + MR_{\text{NC}})(1 - D)}, \quad (3)$$

$$MR_{\text{ratio}} = \frac{RA_{\max} - RA_{\min}}{RA_{\min}} \times 100[\%], \quad (4)$$

where RA_{para} is the parasitic RA, RA_{NOL} is the RA because of the NOL, and RA_{NC} is the RA because of the NCs and is given by $RA_{\text{NC}} = \rho_{\text{NC}} \times t_{\text{NOL}}$. ρ_{NC} is resistivity of NC, t_{NOL} is the thickness of NOL, D is the area density of the NCs, MR_{NOL} is the MR ratio from tunneling effects through the NOL, and MR_{NC} is the MR ratio from the NCs. The observed fitting parameters of MR_{NOL} at $100 \Omega \mu\text{m}^2$ from a parallel circuit model were 15%, 47%, 42%, and 30%, respectively. The maximum at $\text{Co}_{0.7}\text{Fe}_{0.3}$ can be explained using the spin polarization of the tunneling electrons as shown in Ref. 8. The MR_{NC} was estimated to be 1.0%, 4.0%, 5.2%, and 3.5%. The MR ratio from the NCs appears to have a peak at $x=0.5$ in the CoFe alloys. This trend is different from the dependence of the TMR.⁸ This difference between the appearance of the peak in the MR_{NOL} and MR_{NC} in the

CoFe alloys agrees with NCMR, does not depend on the spin polarization of tunneling electrons, and can depend on β . It has been reported that a high annealing temperature was used to obtain a high MR ratio in an NCMR SV.³ Our previous study showed an increase in the MR ratio with annealing at 380°C after annealing at 270°C . The samples of $\text{Co}_{0.5}\text{Fe}_{0.5}$ (circle) showed an increase in the MR ratio from 5.2% to around 11.0% at $0.4 \Omega \mu\text{m}^2$ (shown in Fig. 2). The $\text{Co}_{0.3}\text{Fe}_{0.7}$ (inverted triangle), $\text{Co}_{0.7}\text{Fe}_{0.3}$ (triangle), and $\text{Co}_{0.9}\text{Fe}_{0.1}$ (square) samples showed very little change in the MR ratio after the high temperature annealing process.

To determine the dependence of the MR ratio of the NCs on the ferromagnetic material (in our study the $\text{Co}_{1-x}\text{Fe}_x$ alloy), we investigated the CPP-GMR to determine the value of β for the $\text{Co}_{1-x}\text{Fe}_x$ alloy. Figure 3 shows ΔRA as a function of the thickness of the free ferromagnetic layer of the $\text{Co}_{1-x}\text{Fe}_x$ alloy. ΔRA saturated at increasing thickness for all $\text{Co}_{1-x}\text{Fe}_x$ alloys. The saturation of ΔRA was

FIG. 2. The MR ratio as a function of RA for NCMR samples using a $\text{Co}_{1-x}\text{Fe}_x$ alloy as the ferromagnetic material annealing at a temperature of 270°C (open symbols) and at 270°C annealing followed by 380°C annealing.

FIG. 3. Curves produced by fitting the five-layer VF model to CPP-GMR data with $\text{Co}_{1-x}\text{Fe}_x$ as the ferromagnetic layer.

attributed to the spin diffusion length of the ferromagnet. To evaluate β and γ , we fit the data to the five-layer VF model with the structure of IrMn (semi-infinite)/Co_{1-x}Fe_x (3 nm)/Cu (5 nm)/Co_{1-x}Fe_x t_F/Ru (semi-infinite). We used fitting parameters for the resistivity of $\rho_{\text{Cu}} = 6.5 \mu\Omega \text{ cm}$,¹⁰ $\rho_{\text{Ru}} = 9.5 \mu\Omega \text{ cm}$,¹⁴ $\rho_{\text{IrMn}} = 150 \mu\Omega \text{ cm}$,¹⁵ and the interface resistance between the ferromagnetic and nonmagnetic layers (CoFe/Cu and CoFe/Ru) of $RA_{\text{FM/NM}} = 200 \Omega \text{ nm}^2$.¹⁰ These parameters include the interface between the ferromagnet and IrMn. The spin-diffusion lengths were taken to be $\ell^{\text{Cu}} = 170 \text{ nm}$,¹⁴ $\ell^{\text{Ru}} = 14 \text{ nm}$,¹⁴ and $\ell^{\text{IrMn}} = 1 \text{ nm}$.^{15,16} Although the spin diffusion length of the CoFe alloy was discussed in Ref. 11, in this paper, we fixed all the spin diffusion lengths of the Co_{1-x}Fe_x alloys to be 12 nm. The film resistivity of the CoFe alloy was estimated from the relationship between the ferromagnetic thickness and the sheet conductivity in the CIP geometry. Each film resistivity value for the CoFe alloys is summarized in Fig. 4. With these parameters, we estimated the value of β . Figure 3 shows the fitting curves of the five-layer VF model for Co_{0.9}Fe_{0.1}, Co_{0.7}Fe_{0.3}, Co_{0.5}Fe_{0.5}, and Co_{0.3}Fe_{0.7}. β was found to be 0.61, 0.76, 0.82, and 0.72 for Co_{0.9}Fe_{0.1}, Co_{0.7}Fe_{0.3}, Co_{0.5}Fe_{0.5}, and Co_{0.3}Fe_{0.7}, respectively. Figure 4 shows β as a function of Fe composition in Co_{1-x}Fe_x. There is a peak in β near Co_{0.5}Fe_{0.5}. The dependence of β on Fe composition agrees well with NCMR dependence.

We could only explain the trend that the MR ratio of NCMR SV decreased with decreasing β . However, the experimental MR ratio of NCMR SV (about 11.0%) did not agree with the theoretical MR ratio (over 100%).⁴ We thought the reason that the observed MR ratio was lower than the theoretical MR ratio might be because of oxygen impurities in the NCs.³ Because the NCs were fabricated using an IAO process, a self-organization process created the nano-holes during the oxidization of metal Al. It was difficult to avoid the adsorption of oxygen onto the NCs during the IAO process.

In this final paragraph, we discuss the effect of oxygen impurities in the NCs on both β and the MR ratio of NCM-SV. We fabricated three types of Co_{0.5}Fe_{0.5} films with oxygen impurities, which are FeCoO1, FeCoO2, and FeCoO3 as defined in experimental procedure. The concentration of oxygen impurities was controlled by the thickness of

FIG. 4. Plot showing β and the film resistivity as a function of the concentration of Fe in Co_{1-x}Fe_x alloys.

Co_{0.5}Fe_{0.5}. First, we estimated the resistivity of FeCoO1, FeCoO2, and FeCoO3 using the relation between the ferromagnetic thickness and the sheet conductivity for the CIP geometry of thermally oxidized Si substrate/Ta (3 nm)/Ru (2 nm) FeCo1, 2, or 3 (6 nm, 8 nm, and 10 nm)/Ta (3 nm) structures. The resistivity of films of FeCoO1, FeCoO2, and FeCoO3 was $37.4 \pm 0.53 \mu\Omega \text{ cm}$, $53.6 \pm 5.6 \mu\Omega \text{ cm}$, and $76.7 \pm 8.3 \mu\Omega \text{ cm}$, respectively. When compared with the resistivity of pure Co_{0.5}Fe_{0.5}, it was clear that the presence of oxygen impurities increased the resistivity and that the resistivity of pure Co_{0.5}Fe_{0.5} < FeCoO1 < FeCoO2 < FeCoO3. Next, we estimated the value of β for Co_{0.5}Fe_{0.5} including oxygen impurities using a CPP GMR fit of the five-layer VF model. In this case, we used the fitting parameters of $\gamma = 0.40$ and interface resistance $RA_{\text{FM/NM}} = 200 \Omega \text{ nm}$. The same parameters were used for Co_{0.5}Fe_{0.5}/Cu/Co_{0.5}Fe_{0.5}, because the same interface was used in all samples. Figure 5 shows the fitting curves for the relationship between ΔRA and ferromagnetic thickness of pure Co_{0.5}Fe_{0.5}, FeCoO1, FeCoO2, and FeCoO3. The total free thickness of each sample was controlled by the number of layers of [Co_{0.5}Fe_{0.5}/NO]. β was found to be 0.82, 0.75, 0.65, and 0.35 for pure Co_{0.5}Fe_{0.5}, FeCoO1, FeCoO2, and FeCoO3, respectively. This suggests that an increase in the amount of oxygen impurities decreased β . Despite a smaller value of β for the FeCoO1 sample than β of pure Co_{0.5}Fe_{0.5}, the ΔRA of FeCoO1 sample was larger than for pure Co_{0.5}Fe_{0.5} GMR because of the large resistivity. Figure 6 summarizes the relationship between the resistivity and β for Co_{0.5}Fe_{0.5} with oxygen impurities. Oxygen impurities may act as spin independent scattering sites in the NCs. Spin independent scattering by oxygen impurities would lead to an increase in the resistivity of Co_{0.5}Fe_{0.5} NCs, a decrease in β , and a decrease in the MR ratio of NCMR SVs. According to Fig. 6 and the relationship between β and the theoretical MR ratio of NCMR SVs in Ref. 5, our NCMR SV with a MR ratio of 11.0% can be used to estimate β to be about 0.4 and the resistivity of FeCo NCs to be about $70 \mu\Omega \text{ cm}$ because of oxygen impurities. This estimated resistivity is in reasonable agreement with the previous reports of Refs. 3 and 17. The

FIG. 5. Curves from fits of the five-layer VF model to CPP-GMR data of Co_{0.5}Fe_{0.5} and Co_{0.5}Fe_{0.5} with oxygen impurities using FeCoO1, FeCoO2, and FeCoO3 as the ferromagnetic layer.

FIG. 6. Relationship between the resistivity and β of $\text{Co}_{0.5}\text{Fe}_{0.5}$ with oxygen impurities.

MR ratio of NCMR SVs can be further improved by reducing the oxygen impurities in the NCs.

IV. CONCLUSION

In this paper, we determined the relationship between the MR ratio and the bulk scattering spin asymmetry coefficient of the ferromagnet, β , using the ferromagnetic material dependence of the MR ratio on different NCMR SVs. The reason for a lower experimental MR ratio compared with the theoretical MR ratio is because of the decrease in β caused by oxygen impurities. Unlike the dependence of TMR on a particular CoFe alloy, the peak of the MR in the NCMR SVs appears at $\text{Co}_{0.5}\text{Fe}_{0.5}$. Furthermore, we clarified the relation between the spin asymmetry, β , and the Fe composition in $\text{Co}_{1-x}\text{Fe}_x$ using a CPP-GMR fit with a five-layer VF model. The maximum in β was near $\text{Co}_{0.5}\text{Fe}_{0.5}$. The difference between the experimental MR ratio and the theoretical MR ratio was caused by spin independent scattering from the oxygen impurities. The reason for this was determined by

estimating β in the sample where oxygen impurities were artificially introduced. Increasing the amount of oxygen impurities in CoFe increased the resistivity and decreased β . The NCs of our NCMR SVs might contain oxygen impurities. The MR ratio of NCMR SVs can be improved by reducing the concentration of oxygen impurities in NCs.

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