## Inverse Tunnel Magnetocapacitance in Fe/Al-oxide/Fe<sub>3</sub>O<sub>4</sub>

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Magnetocapacitance (MC) effect in spintronic devices has attracted much interest due to their fascinating spin-based phenomena, such as spin capacitance, frequency-dependent spin transport, and potential applications as sensitive magnetic sensors, high-frequency devices, and energy storage materials. The MC effect has been observed in magnetic tunnel junctions (MTJs) [1–3], molecular spin valve devices [4], magnetic nanogranular films [5], and magnetic single-electron transistors [6]. The MC in MTJs is generally referred to as tunnel magnetocapacitance (TMC). In normal TMC, the capacitance  $C_P$  is high for the parallel (P) configuration of the magnetization vectors in both ferromagnetic layers adjacent to the barrier, and  $C_{AP}$  is low for the antiparallel (AP) configuration. In this current work, we report an *inverse TMC* effect (i.e.,  $C_P < C_{AP}$ ) for the first time in Fe/AlO<sub>x</sub>/Fe<sub>3</sub>O<sub>4</sub> MTJs.

We prepared the device structure by using a molecular beam epitaxy system [7] with a base pressure of  $10^{-8}$  Pa. The MTJ stack consists of MgO(110)/MgO(20 nm)/NiO(5 nm)/Fe<sub>3</sub>O<sub>4</sub>(60 nm)/AlO<sub>x</sub>(2–4 nm)/Fe(10 nm)/Au(30 nm). We patterned the MTJ structures with a junction area of  $10 \times 10 \ \mu\text{m}^2$  by using standard photolithography with Ar ion-milling and SiO<sub>2</sub> insulation overlayer. As shown in Fig. 1, the inverse TMC effect was clearly observed in Fe/AlO<sub>x</sub>/Fe<sub>3</sub>O<sub>4</sub> MTJs. Fig. 2 shows the frequency dependence of inverse TMC and TMR. The magnitude of the inverse TMC reaches up to 11.4% at room temperature. Excellent agreement between theory and experiment is achieved for the entire applied frequency range using Debye-Fröhlich model (combined with Zhang formula and parabolic barrier approximation) and spin-dependent drift-diffusion model. Furthermore, our theoretical calculations predict that the inverse TMC effect could potentially reach 150% in MTJs with a positive and negative spin polarization of 65% and -42%, respectively, as shown in Fig. 3. These theoretical and experimental findings provide a new insight into both static and dynamic spin-dependent transports. They will open broader opportunities for device applications, such as magnetic logic circuits and multi-valued memory devices. This research was supported by JSPS KAKENHI (No. 15H03981), MEXT Dynamic Alliance, CSRN at Tohoku University, CRP of ICR at Kyoto University (No. 2016-57), and NSF at Brown University (No. DMR-1307056).



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