Indium oxide and indium-tin-oxide channel ferroelectric gate thin film transistors with yttrium doped hafnium-zirconium dioxide gate insulator prepared by chemical solution process

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Ferroelectric gate transistor (FGT) with yttrium doped hafnium-zirconium dioxide (Y-HZO) gate insulator and oxide channel with various thicknesses of In2O3 and ITO were fabricated by chemical solution deposition. First, ferroelectric properties of Y-HZO in the metal-ferroelectric-semiconductor structure with 5–22 nm thick In2O3 and 6–24 nm thick ITO, have been confirmed by polarization–voltage and capacitance–voltage (C–V) characteristics. The C–V curves showed clear butterfly loops showing the depletion of In2O3 and ITO layer. Secondly, the device performance of FGTs has been evaluated with various thicknesses of In2O3 and ITO channel layer. The fabricated FGTs exhibited typical n-channel transistor operation with a counterclockwise hysteresis loop due to the ferroelectric nature of the Y-HZO-gate insulator. It was found that FGT shows a low subthreshold voltage swing, high on/off drain current ratio of 106, large on current, and memory window.

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1. Introduction

Ferroelectric gate transistor (FGT) has gained much attention due to low power consumption, high endurance, and non-volatile function of the ferroelectric material used as a gate insulator.1,2) Silicon channel based ferroelectric field-effect transistors have been most intensively studied for ferroelectric random-access memory applications.3–5) However, Si channel FGTs have several issues to be considered. Firstly, an interfacial SiO2 layer is formed between the Si channel and the ferroelectric layer during the HfO2 deposition and annealing process, which reduces the voltage applied to the ferroelectric layer and sometimes exhibits poor electrical properties.6) Secondly, the channel charge of Si-MOSFET does not match the ferroelectric polarization (charge mismatch problem). Most of the ferroelectric materials have spontaneous remnant polarization of 10–30 μC cm−2, while the charge required to control the channel conductivity of MOSFET is usually less than 1 μC cm−2. Hence, only minor loops of the ferroelectric material are used in Si-channel FGT, which reduces the memory window (MW) (threshold voltage shift).7) One method to avoid the charge mismatch problem in FGT is to use an oxide channel.8) We have previously demonstrated that full polarization of ferroelectric gate insulator can be utilized in oxide channel FGTs.

In our previous works, conventional ferroelectric materials such as Pb(Zr, Ti)O3 (PZT) and (Bi, La)4Ti3O12 (BLT) were used to fabricate FGTs.8–10) However conventional materials have several problems such as relatively small bandgap (e.g. 3–4 eV), poor compatibility with CMOS technology, and scaling effect has to be taken into consideration.1,17,18) Recently HfO2 based thin films witnessed significant interest as complementary metal-oxide-semiconductor (CMOS) compatible ferroelectrics since the first report of ferroelectricity in HfO2 in 2011.19) HfO2 based ferroelectric thin films have several advantages over conventional ferroelectric materials. Firstly, HfO2 has a wide bandgap which can effectively suppress the leakage current.20) Secondly, HfO2 exhibits ferroelectricity even in ultra-thin films (1–5 nm).20) Thirdly, ferroelectric HfO2 films are highly compatible with CMOS technology.21) Therefore, ferroelectric HfO2 films are a promising candidate for large scale integration.22)

As reported previously, the orthorhombic phase (hereafter called O-phase) is responsible for ferroelectricity in HfO2 films.22) O-Phase can be stabilized by doping with other elements such as Si,23) Zr,24) Y,24) Al,25) and La.26) In particular, Zr doped HfO2 or Hf1-zZrO2 (HZO) has gained great attention due to ferroelectricity in wide range Zr composition unlike the other dopants and large remnant polarization 20 μC cm−2. Therefore, HZO can be a promising candidate for oxide channel FGTs applications. However, in contrast to the large number of reports in Si-based FGTs, there are only a few reports on oxide-channel FGT with HfO2-based ferroelectric materials.27–29) In these reports, two things are common, firstly HZO films were deposited by atomic layer deposition, and secondly, the oxide channel layer was deposited by sputtering. In this work, we prepared both HZO gate insulator and oxide channel by chemical solution deposition (CSD). There is no report on HZO based oxide channel FGTs prepared by CSD.

Previously, we reported electrical properties of the metal-ferroelectric-semiconductor (MFS) structure using CSD yttrium doped HZO (Y-HZO) annealed in oxygen and CSD indium-tin-oxide (ITO).30) It was found that the C–V curves showed a clear butterfly-shaped hysteresis due to the ferroelectricity with a decrease in capacitance due to the depletion of ITO. Additionally, we reported P–E loops and leakage current of CSD Y-HZO films can be improved drastically by a vacuum annealing process.31) Therefore, we fabricated MFS structures using vacuum annealed CSD Y-HZO films and 13 nm thick CSD ITO channel layer and reported ferroelectric properties along with the transistor operation in SSDM 2020.32) In this paper, we report bottom gate FGTs.
using CSD Y-HZO as a gate insulator and CSD indium oxide (In$_2$O$_3$) or indium-tin-oxide (ITO) with thicknesses of 5–22 nm and 6–24 nm, respectively. One of the reasons for using In$_2$O$_3$ or ITO channel with a high carrier concentration of $10^{19}$ or $10^{20}$ cm$^{-3}$, is to demonstrate the “ferroelectric” Y-HZO gate insulator clearly. As we previously reported, a thin ITO channel can be depleted completely due to the large charge controllability of the ferroelectric gate insulator. Since the ferroelectric O-phase in HfO$_2$-based material is metastable, observing transistor behavior of ITO (or In$_2$O$_3$)/Y-HZO FGTs would be an indicator of ferroelectricity of Y-HZO gate insulator after the device fabrication. Reliability issues of the ITO (or In$_2$O$_3$)/Y-HZO FGTs fabricated by CSD will be the subject of future investigation.

2. Experimental details

Y-HZO films were fabricated by the procedure we reported previously. Platinized silicon substrates (Pt/Ti/SiO$_2$/Si) with dimensions of 1.5 × 1.5 cm$^2$ were used to deposit thin films by CSD. Precursor solution for Y-HZO was prepared by using hafnium (IV) acetylacetonate [Hf(acac)$_4$], zirconium (IV) acetylacetone [Zr (acac)$_4$], and yttrium (III) acetylacetone [Y(acac)$_3$] as starting materials. We prepare the source solution in air without using a glove box.

![Fig. 1. (Color online) (a) Process flow of FGT (b) In$_2$O$_3$ and ITO film thickness as a function of source solution concentration, (c) schematic illustration of Pt/In$_2$O$_3$ or ITO/Y-HZO/Pt MFS structure (d) schematic illustration of FGT (e) schematic of FGT (cross section).](image)

We spin-coated a platinized substrate, followed by drying at 225 °C for 3 min in air. The crystallization was done by using rapid thermal annealing at 800 °C for 3 min in a vacuum environment. The thickness of Y-HZO films was 33 nm. In$_2$O$_3$ and ITO source solution with various solution concentrations was prepared by mixing In(acac)$_3$ in PrA and In(acac)$_3$ and Sn(acac)$_2$ in PrA, respectively, at 120 °C for 1 h. The thickness of In$_2$O$_3$ and ITO was controlled by changing precursor solution concentration as shown in Fig. 1(b). The thickness of In$_2$O$_3$ was 5, 12, 22, 39 and 57 nm for the source solution concentration of 0.025, 0.05, 0.10, 0.20 and 0.30 mol kg$^{-1}$, respectively, while the thickness of ITO was 6, 13, 24, 40 and 58 nm for 0.025, 0.05, 0.10, 0.20 and 0.30 mol kg$^{-1}$, respectively. As shown in Fig. 1(b), the thickness of In$_2$O$_3$ and ITO are about the same and we use In$_2$O$_3$ and ITO layer whose thickness is less than 24 nm for FGT fabrication.

Precursor solution of In$_2$O$_3$ and ITO with a concentration of 0.025–0.10 mol kg$^{-1}$ was spin-coated on Y-HZO films, followed by drying at 100 °C for 3 min in air. Then, the samples were annealed at 600 °C for 15 min in an O$_2$ environment. Pt source and drain electrodes were deposited by sputtering and patterned by the lift-off process. Next, the device region was isolated by wet etching. The channel length and width are 5 and 100 μm, respectively. Finally, the bottom gate electrode is accessed by photolithography and etching. The fabricated FGTs structure is shown in Figs. 1(d) and 1(e). Pt/ITO/Y-HZO/Pt capacitors were also fabricated to measure the ferroelectric properties of the MFS structures as shown in Fig. 1(c).

Film thickness was measured by step profilometer (AlphaStep-D500 by KLA tencor). The crystallinity of films was identified by using X-ray diffraction (XRD; X’Pert PRO MRD Epi from PANalytical). Polarization–voltage field (P–V) with current–electric field (I–V) characteristics of all
samples were measured by a ferroelectric test system (TOYO Corporation Model FCE-1) at 1 kHz whereas capacitance–voltage (C–V) measurements were achieved by using a precision component analyzer (Wayne Kerr 6440B Model) at 10 kHz. The current density–voltage (J–V) curves, output and transfer characteristics of FGT were acquired with a semiconductor parameter analyzer (Agilent 4155B).

3. Result and discussion

3.1. Crystallinity of films

XRD pattern of 5% Y-HZO annealed in a vacuum environment at 800 °C is shown in Fig. 2. The diffraction peak around 30.5° and 35.4° were observed. The most intense peak was observed at 30.5 °C which corresponds to 111 of O-phase or cubic phase (C-phase) or tetragonal phase (T-phase). A sub-peak around 35.2° reflects 200 of O-phase, C-phase or T-phase. It is worth noting that negligible monoclinic phase was observed in the XRD pattern of CSD Y-HZO films.

3.2. Electrical properties of MFS structures

3.2.1. In2O3/Y-HZO MFS structure.

In order to confirm the ferroelectricity of the Y-HZO films, the P–V and C–V characteristics were measured for the In2O3/Y-HZO MFS structures with various thicknesses of In2O3. Since the thin...
In$_2$O$_3$ layer may be depleted in the MFS structure, we use voltage instead of the electric field as a horizontal axis. Figures 3(a), 3(c), and 3(e) shows $P$–$V$ loops, and Figs. 3(b), 3(d), and 3(f) show the corresponding current response of Y-HZO films with an In$_2$O$_3$ thickness of 5, 12, and 22 nm, respectively. As shown in Figs. 3(a) and 3(b), the $P$–$V$ loops with 5 nm thick In$_2$O$_3$ show clear ferroelectric hysteresis loops with a corresponding switching current response. The $P$–$V$ loops of the MFS structure with 12 nm thick In$_2$O$_3$ also show clear ferroelectric hysteresis loops with a slight increase in saturated polarization and switching current response. When the In$_2$O$_3$ thickness was 22 nm, as shown in Fig. 3(e), $P$–$V$ loops become slightly rounded, which is probably due to an increase of leakage current as shown later in Fig. 4. Figure 3(e) shows a corresponding switching current where switching current can be observed.

Figure 4 shows the leakage current density of the In$_2$O$_3$/Y-HZO MFS structures with 5 nm (dotted line) and 22 nm (solid line) thick In$_2$O$_3$. Leakage current at 1 MV cm$^{-1}$ for the In$_2$O$_3$/Y-HZO MFS structure with 5 nm (dotted line) and 22 nm (solid line) thick In$_2$O$_3$ are 7.4 $\times$ 10$^{-6}$ and 1.2 $\times$ 10$^{-4}$ A cm$^{-2}$, respectively. The increase in leakage current when the thickness of In$_2$O$_3$ increased to 22 nm is consistent with the rounded $P$–$V$ loops observed in Fig. 3(e). In$_2$O$_3$/Y-HZO MFS structure with 12 nm In$_2$O$_3$ shows similar leakage current density of Y-HZO films with 5 nm thick In$_2$O$_3$ (not shown in the figure). In addition, remnant polarization ($P_r$) of Y-HZO films with different sweep voltage for different thicknesses of In$_2$O$_3$ is plotted in Fig. 5. It was found that Y-HZO film with In$_2$O$_3$ thickness of 5 and 12 nm shows good saturation of $P_r$, as observed in other HfO$_2$ based ferroelectric films. In contrast, Y-HZO films with an In$_2$O$_3$ thickness of 22 nm show no saturation of $P_r$, probably due to the increase of leakage current. Remnant polarization $P_r$ and coercive field ($E_c$) of Y-HZO films with different thicknesses of In$_2$O$_3$ deduced from Figs. 3(a), 3(c) and 3(e), are approximately 8.7 $\mu$C cm$^{-2}$ and 1.3 MV cm$^{-1}$, 9.3 $\mu$C cm$^{-2}$ and 1.3 MV cm$^{-1}$, 14.3 $\mu$C cm$^{-2}$ and 2.1 MV cm$^{-1}$, respectively.

In addition to $P$–$V$ loops, $C$–$V$ measurement was carried out to confirm the ferroelectric nature of Y-HZO with different thicknesses of In$_2$O$_3$. Figures 6(a)–6(c) shows $C$–$V$ curves of the In$_2$O$_3$/Y-HZO MFS structures with an In$_2$O$_3$ thickness of 5, 12, and 22 nm, respectively. All samples show clear butterfly-shaped loops with a decrease in capacitance on the positive side, which demonstrates ferroelectric properties of Y-HZO films clearly. Since positive voltage is applied to the top electrode, the observed decrease in capacitance value is due to the depletion of In$_2$O$_3$. A MW, width of the hysteresis loop in $C$–$V$ curve which relates to the threshold voltage shift in the device, was estimated for a sweep voltage of $\pm$ 8 V from Figs. 6(a)–6(c), and these are approximately 1.9 V, 3.6 V and 3.9 V, respectively.

### 3.2.2. ITO/Y-HZO MFS structure.

As discussed in the previous section, $P$–$V$ and $C$–$V$ measurements were performed to verify the ferroelectric nature of Y-HZO with In$_2$O$_3$, and similar measurements were carried out for ITO/Y-HZO MFS structures with different thickness of ITO. Figures 7(a), 7(c), and 7(e) show $P$–$V$ loops, and Figs. 7(b), 7(d), and 7(f) show $P$–$V$ corresponding current response of ITO/Y-HZO MFS structures with 6, 13, and 24 nm thick ITO, respectively. $P$–$V$ loops of ITO/Y-HZO MFS structure with 6 nm thick ITO shows clear hysteresis loops and $I$–$V$ loops show clear switching response as shown in Figs. 7(a) and 7(b), respectively. Likewise, $P$–$V$ and $I$–$V$ loops of ITO/Y-HZO MFS structures with 13 nm thick ITO show clear ferroelectric hysteresis loop and clear switching response as depicted in Figs. 7(c) and 7(d). When the thickness of the ITO layer was increased to 24 nm, $P$–$V$ loops of ITO/Y-HZO MFS structure become slightly rounded due to an increase in leakage current, while the clear switching response still can be observed as shown in Fig. 7(f). This is similar to the trend observed for the In$_2$O$_3$/Y-HZO MFS structures.

Figure 8 shows the leakage current density of ITO/Y-HZO MFS structure with 6 nm (dotted lines) and 24 nm (solid line) thick ITO layer. The leakage current at 1 MV cm$^{-1}$ for Y-HZO films with 6 and 24 nm thick ITO layer deduced from Fig. 8 are 1.34 $\times$ 10$^{-6}$ and 1.2 $\times$ 10$^{-4}$ A cm$^{-2}$, respectively, which suggests that leakage current was increased when the...
thickness of the ITO layer is 24 nm. ITO/Y-HZO MFS structure with 13 nm ITO shows similar leakage current density of ITO/Y-HZO MFS structure with 6 nm thick ITO. Hu et al., reported diffusion of In into HfO2 film from InAs substrate. Similar tendency for the MFS structure with thicker ITO or In2O3 could be possible which would cause the increase of leakage current, but further investigation is required to clarify the reason for the increase of leakage current for the MFS structure with relatively thick In2O3 or ITO layer.

In addition, remnant polarization (Pr) of Y-HZO films as a function of sweep voltage for the ITO/Y-HZO MFS structures with different thicknesses of the ITO layer is shown in Fig. 9. It was found that MFS structures with 6 and 13 nm thick ITO show good saturation of Pr. On the other hand, the saturation of the Pr is not good for the MFS structures with 24 nm thick ITO layer due to an increase of leakage current.

In addition to P–V loops, we evaluated C–V characteristics for ITO/Y-HZO MFS structures, too. Figures 10(a)–10(c) show C–V curves of Y-HZO films with a thickness of ITO 6, 13, and 24 nm, respectively. All samples depict clear butterfly-shaped loops with a decrease in capacitance on the positive side. The observed decrease in capacitance value is due to the depletion of ITO. The MW of Y-HZO films with 6, 13, and 24 nm thick ITO layer estimated for a sweep voltage of ±8 V in Figs. 5(a)–5(c), are approximately 2.2 V, 3.7 V, and 4.7 V, respectively.

3.3. FGT device performance with different thickness of oxide channel

3.3.1. FGT with CSD In2O3 channel. Since we confirmed good ferroelectric properties of the Y-HZO films using the MFS structure, next we fabricated transistors. Figure 11 shows the electrical properties of FGT with a different channel thickness of In2O3 with output and transfer characteristics. Figures 11(a), 11(c), and 11(e) show $I_D$–$V_D$ characteristics of FGT with In2O3 channel thickness of 5 nm, 12 nm, and 22 nm, respectively. $I_D$–$V_D$ characteristics show typical n-channel field-effect transistor with linear behavior in the drain current at a low drain voltage, and at a high drain voltage, good saturation of the drain current for all samples. The channel length L and width W are 5 and 100 μm, respectively. A large saturated on current of approximately 8 mA (0.08 mA μm$^{-1}$) at $V_G$ 6 V, 11 mA (0.11 mA μm$^{-1}$) at $V_G$ 3 V, and 11 mA (0.11 mA μm$^{-1}$) at $V_G$ 3 V were obtained for channel thickness of 5, 12, and 22 nm, respectively. Figures 11(b), 11(d), and 11(f) show $I_D$–$V_G$ characteristics of FGT with In2O3 channel thickness of 5, 12, and 22 nm, respectively. $I_D$–$V_G$ characteristics show a typical n-channel transfer curve for all samples with a counterclockwise hysteresis loop due to the ferroelectric nature of the Y-HZO gate insulator. FGT performance parameter such as on/off drain current ratio, subthreshold voltage swing (SS), and MW estimated from Fig. 6.
Figs. 11(b), 11(d), and 11(f), are approximately $7 \times 10^6$, 70 mV/dec and 0.66 V; $6.8 \times 10^6$, 172 mV/dec and 3.1 V; $1 \times 10^6$, 221 mV/dec and 3.4 V for channel thickness of 5, 12, and 22 nm, respectively. The SS increases with increasing channel thickness which is consistent with the previous report.\textsuperscript{27} The SS value as low as 70 mV/dec and on/off ratio as high as $7 \times 10^6$ was obtained for channel thickness of 5 nm which is comparable to the best values reported so far.\textsuperscript{27,29} The MW estimated from transfer characteristics shows good agreement with the MW estimated from $C$–$V$ curves. However, in the ideal case, the theoretical MW is given by $2E_Cd$ or $2V_C$, where $d$ is the thickness of the ferroelectric film, $E_C$ is the coercive field, and $V_C$ is the coercive voltage.\textsuperscript{8} $2V_C$ values estimated from the $P$–$V$ loop at ±8 V for Y-HZO films with different thicknesses of In$_2$O$_3$ shown in Figs. 3(a), 3(c), and 3(e), are approximately 6.2 V, 6.6 V and 7.1 V, respectively. The MW values from $C$–$V$
curves and transfer characteristics are lower than $2V_C$, which might be due to the co-existence of charge injection which reduces the MW especially for the device with a 5 nm thick In$_2$O$_3$ channel.

3.3.2. FGT with CSD ITO channel. Next, FGT with CSD ITO channel whose thickness is 6, 13, and 24 nm were also characterized. Figure 12 shows the output and transfer characteristics of FGTs with different channel thickness of ITO. Figures 12(a), 12(c) and 12(e) show $I_D$–$V_D$ characteristics of FGT with In$_2$O$_3$ channel thickness of 6, 13, and 24 nm, respectively. $I_D$–$V_D$ characteristics clearly demonstrate standard n-channel MOSFET operation with good saturation of the drain current for all samples. Note that a large saturated on drain current was obtained. The on current estimated from Figs. 12(a), 12(c), and 12(e), are approximately 3 mA (0.03 mA μm$^{-1}$) at $V_G$ 6 V, 16 mA (0.16 mA μm$^{-1}$) at $V_G$ 3 V, and 18 mA (0.18 mA μm$^{-1}$) at $V_G$ 3 V for the FGTs with channel thickness of 6, 13, and 24 nm, respectively. Figures 12(b), 12(d) and 12(f) shows $I_D$–$V_G$ characteristics of FGT with ITO channel thickness of 6, 13, and 24 nm, respectively. $I_D$–$V_G$ characteristics clearly exhibit a standard n-channel transfer curve with a counterclockwise hysteresis loop due to the ferroelectric nature of the Y-HZO. In the case of 6 nm thick ITO channel, the clockwise hysteresis was observed for the large gate voltage region, which is probably due to the charge injection which was also observed in 1 nm thick ITO channel FGTs reported previously. On/off drain current ratio, SS, and MW estimated from Figs. 12(b), 12(d), and 12(f), are approximately $5.8 \times 10^6$, 117 mV/dec and 1.1 V; $5.1 \times 10^6$, 198 mV/dec and 3.9 V; $1 \times 10^6$, 290 mV/dec and 4.5 V, for the FGTs with channel thickness of 6, 13, and 24 nm, respectively. The MW estimated from C–V curves shows good agreement with MW estimated from transfer characteristics. However, the MW values from transfer characteristics and CV curves are lower than $2V_C$ for the ITO channel devices, which is similar to the FGT with In$_2$O$_3$ channel. 2 $V_C$ values estimated from the $P$–$V$ loop at ±8 V of Figs. 7(a), 7(c), 7(e) are as large as 6 V, regardless of the ITO thickness. This indicates the charge injection co-exists and the charge injection effect is more serious.
when the ITO thickness is as thin as 6 nm. FGT with 6 nm thick ITO channel shows low SS value but charge injection is dominant as result clockwise hysteresis as observed at higher voltage and the MW decreases. When the channel thickness is increased to 24 nm, the off current and SS become large. Hence, the FGT with channel thickness around 13 nm shows comparatively good electrical properties with low SS value, high on/off drain current ratio and large MW.

4. Conclusion

We have fabricated bottom gated FGT with Y-HZO gate insulator and 5–24 nm thick In$_2$O$_3$ and ITO as a channel. At first, the ferroelectric properties of Y-HZO with different thicknesses of In$_2$O$_3$ and ITO were evaluated. It was found that Y-HZO films show good ferroelectric properties when the thickness of In$_2$O$_3$ and ITO was less than 13 nm. When the thickness of In$_2$O$_3$ and ITO increased to 22–24 nm, the MFS structures showed ferroelectric nature but $P$–$V$ loops became slightly rounded due to the increase of leakage current. Fabricated bottom gate FGTs with thin In$_2$O$_3$ and ITO channel, demonstrated normal n-channel transistor operation with hysteresis in the transfer curves due to the ferroelectric nature of the Y-HZO gate insulator. A low SS value of 70 mV/decade (5 nm In$_2$O$_3$ channel), high on/off ratio of $10^6$, large MW, and large on current were obtained and the FGT with a channel thickness around 13 nm shows better device performance.

Fig. 11. (Color online) Electrical properties of FGT with Y-HZO gate insulator with different thickness of In$_2$O$_3$ channel. (a) $I_D$–$V_G$ and (b) $I_D$–$V_D$ characteristics channel thickness of 5 nm, (c) $I_D$–$V_D$, and (d) $I_D$–$V_G$ characteristics of the FGT with channel thickness 12 nm, (e) $I_D$–$V_D$, (f) $I_D$–$V_G$ characteristics of the FGT with channel thickness 22 nm.
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Fig. 12. (Color online) Electrical properties of FGT with Y-HZO gate insulator with different thickness of ITO channel, (a) $I_D$–$V_D$ and (b) $I_D$–$V_G$ characteristics of the FGT with channel thickness of 6 nm, (c) $I_D$–$V_D$ and (d) $I_D$–$V_G$ characteristics of the FGT with channel thickness 13 nm, (e) $I_D$–$V_D$ and (f) $I_D$–$V_G$ characteristics of the FGT with channel thickness 24 nm.

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