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Ultrahigh Energy Density due to Self-growing Double Dielectric Layers at Titanium/Sol-gel-derived Amorphous Aluminium Oxide Interface

Zhen Su, Manwen Yao*, Xi Yao

Functional Materials Research Laboratory, School of Materials Science & Engineering, Tongji University, No. 4800 Cao'an Road, Shanghai 201804, China
E-mail: yaomw@tongji.edu.cn, 18901998729@126.com

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1 The TEM diffraction photographs of TO and AO/TO layers

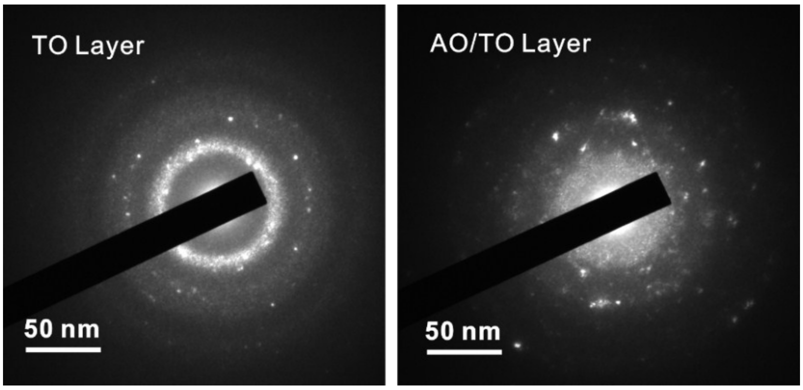

Fig. S1 TEM diffraction photographs of TO layer and AO/TO layer

2. The preparation of (AO/TO)/AmAO and 24-nm-TO/(AO/TO)/AmAO

As shown in Fig. 3a in the manuscript, the current density is divided into two phases when the positive voltage was applied again: (a) < 90 MV m^{-1}, (b) > 90 MV m^{-1}. The two phases mean two layers are developed. The current density at < 90 MV m^{-1} is lower than that at > 90 MV m^{-1}. It is suggested that the Al_2O_3/TiO_x layer is developed < 90 MV m^{-1} because the resistivity of Al_2O_3/TiO_2 layer is higher than that of TiO_2 layer. We prepared ~15-nm-thick Ti electrode by magnetron sputtering at 50 w for 200 s. The 60 nm Au electrode was sputtered on the samples, and then applied electric field of 90 MV m^{-1} (~23 V) and 500 MV m^{-1} (~140 V), respectively.

3 The transport behavior of titanium ions

As shown in Fig. 6, the thickness of titanium electrode decreased from 65 to 44 nm. If the titanium transformed into titanium dioxide (TO), the thickness ratio should be:

$$\frac{d_{TO}}{d_{Ti}} = \frac{M_{TO}\rho_{Ti}}{2M_{Ti}\rho_{TO}} = 1.9 \qquad (1)$$

where M_{TO} is the molar mass of titanium dioxide (79.9 g mol^{-1}); M_{Ti} is the molar mass of titanium (47.9 g mol^{-1}); ρ_{Ti} is the density of titanium (4.5 g cm^{-3}); ρ_{TO} is the density of titanium dioxide (3.9 g cm^{-3}).

In practice, the ratio is:

$$\frac{d_{TO}}{d_{Ti}} = \frac{34\ \text{nm}}{(65 - 44)\ \text{nm}} = 1.6 \qquad (2)$$

The experimental value was lower than the theoretical value confirming ions transported into newly-formed aluminum oxide layer.