Experimental and Theoretical Studies of Hydroxyl-Induced Magnetism in TiO Nanoclusters

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I. Cluster deposition

Clusters are fabricated from inert-gas condensation with a cluster deposition system. Figure S1 shows a schematic diagram of the cluster deposition system. As can be seen, it is comprised of two parts, the gas condensation chamber and the deposition chamber. Ti oxide clusters can be fabricated from two different ways via *in-situ* and *ex-situ* oxidation. *In-situ* oxidation is realized by feeding oxygen to the condensation chamber. By varying the oxygen partial pressure clusters of various compositions and phases can be obtained. *Ex-situ* cluster deposition is realized by annealing as-produced Ti clusters in an oxygen atmosphere.

(i) Cluster deposition via *in-situ* oxidation

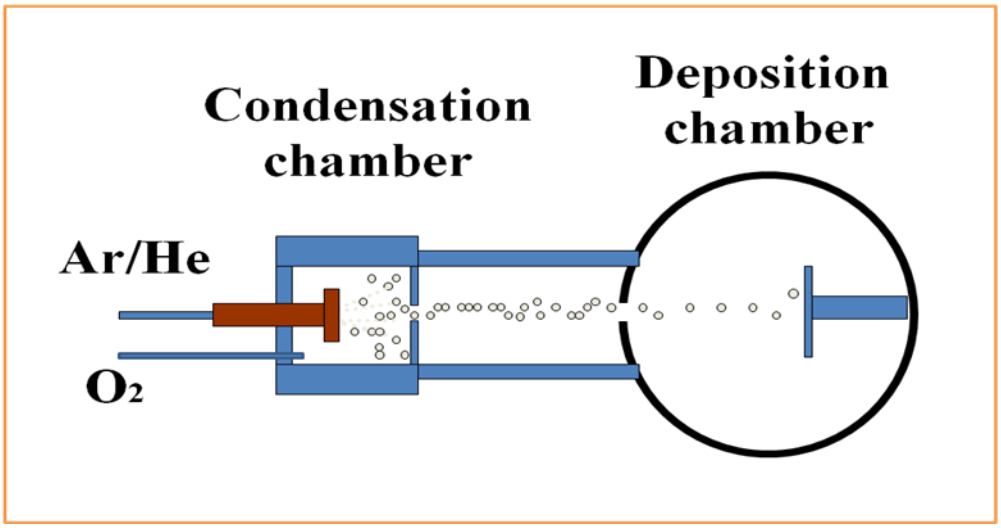

Figure S1. Cluster deposition system. *In-situ* oxidation is realized through feeding oxygen to the condensation chamber.

(ii) Cluster deposition via *ex-situ* oxidation

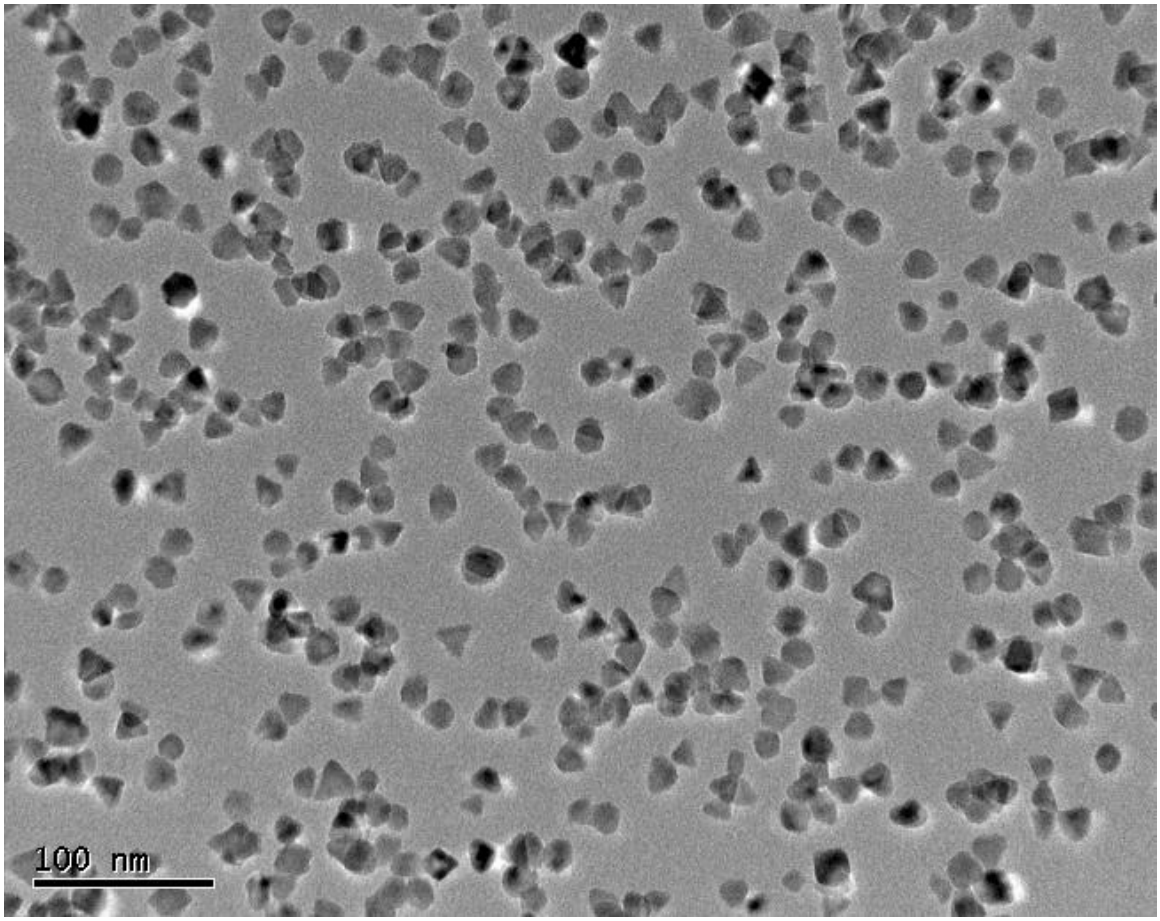

Figure S2. TEM image of as-produced Ti clusters for *ex-situ* oxidation.

II. Comparative study of the aging behavior of TiO and TiO_2 clusters in ambient air.

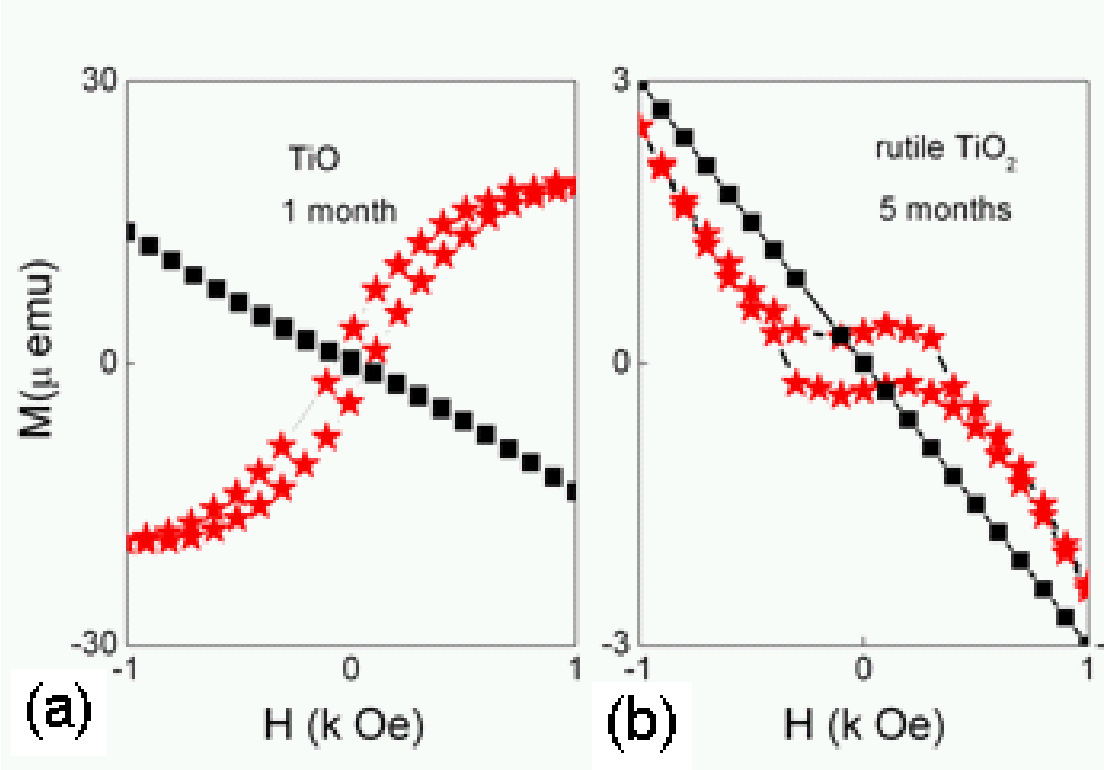

Figure S3. Comparison of the aging rates of various Ti oxide clusters in ambient air. (a) Hysteresis loops for TiO clusters after deposition (black squares) and after aging in air for one month (red stars). (b) Hysteresis loops for rutile TiO_2 clusters after deposition (black squares) and after aging in air for 5 months (red stars). The Si substrate background is included to retain the original shape of the hysteresis loops. The two samples have comparable thicknesses. In comparison, the magnetization of the TiO clusters is roughly an order of magnitude higher than that of the rutile TiO_2 clusters.

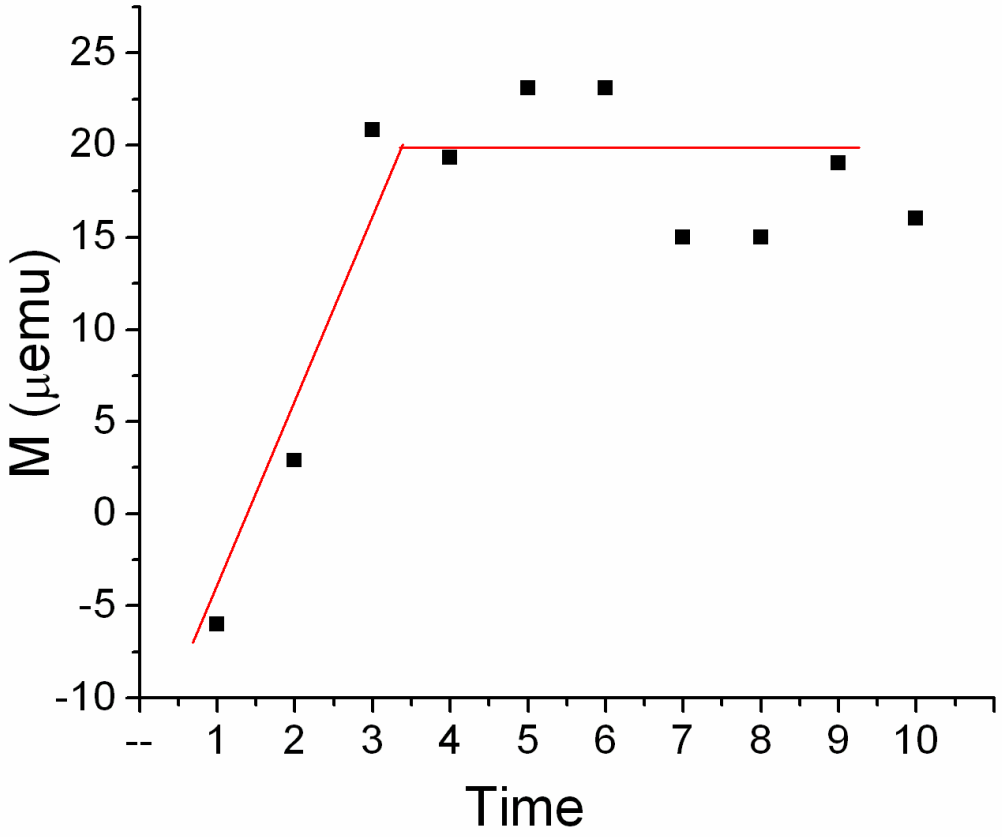

Figure S4. Variation of the magnetic moment as a function of storage time in moist air for a thinner TiO sample with a thickness of 40 nm. The same equilibrium magnetization is obtained as in the thicker sample.

III. Possible generality of the hydroxyl-induced magnetic moment for other room-temperature-ferromagnetic oxides from the literature.

Moment relaxations induced by oxygen annealing: comparison of TiO to other RTFO from literature.

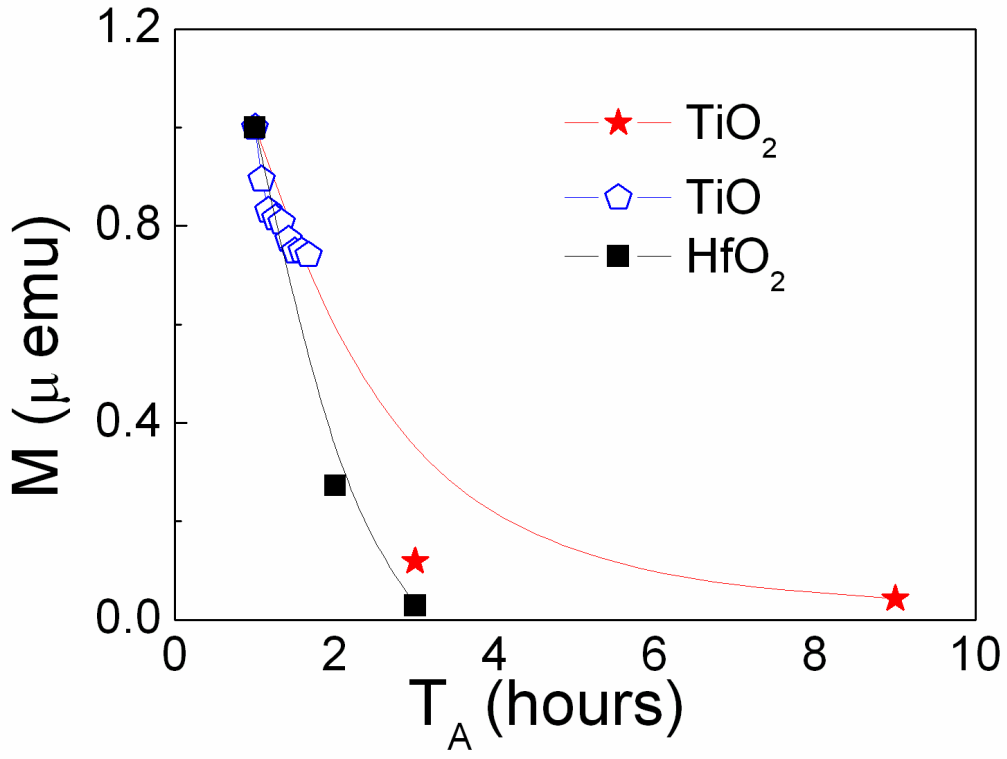

Figure S5. Magnetization relaxation during oxygen annealing: TiO clusters vs. other oxides from literature. [1]

References

[1]. N. Hong, J. Sakai, N. Poirot and V. Brizé, *Phys. Rev. B* 2006, **73**, 132404.