Supporting Information

Hole Carriers Doping Effect on the Metal-Insulator Transition of N-Incorporated Vanadium Dioxide Thin Films

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Regarding the homogeneity of the doped VO_2 films, it should be no problem for the in-plane doping of the film given the larger ion beam size in contrast with the 10 mm x 10 mm sample. While out of the plane (along the normal direction), the ion implantation method for material doping is characterized with a controllable dopant distribution in the matrix material. The distribution can be simulated by the stopping and range of ions in matter (SRIM) code depending on the ionic energy and dosage. For the present experiment, we simulate the N_2^+ distribution at 2 keV and 1.2 μA/cm^2 in VO_2 film and the result is shown in Figure R1. It can be seen the maximum concentration is at the depth of 4.8 nm although the doped N can reach furthest the depth of ~12 nm.

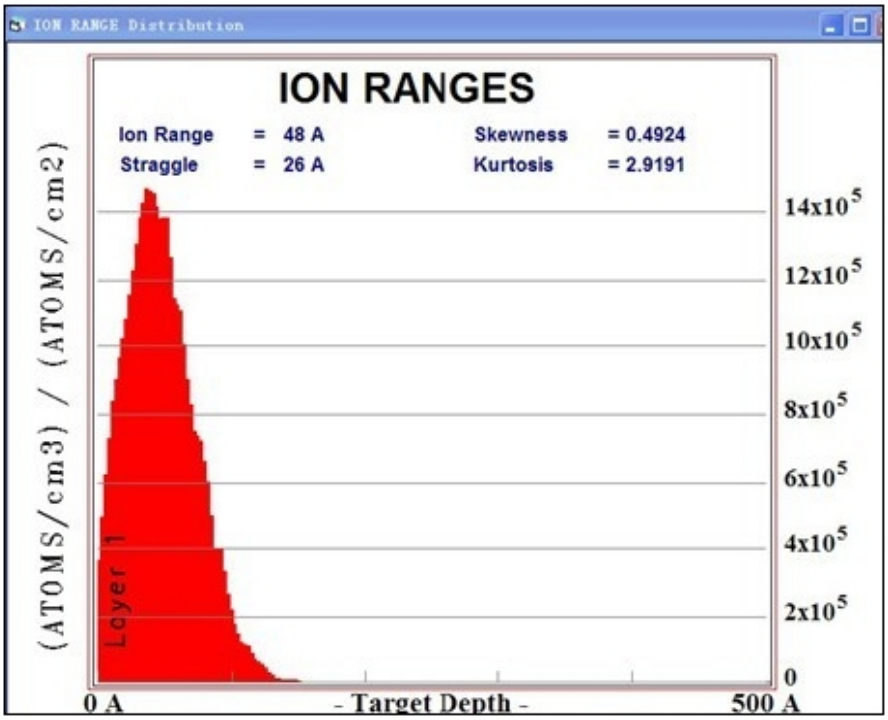

Fig. R1 Simulation of the N_2^+ distribution in VO_2 film. The ionic energy is 2 keV and dosage is 1.2 μA/cm^2. The maximum concentration is at the depth of 4.8 nm although the doped N can reach furthest the depth of ~12 nm.

In order to account for the nitrogen distribution across the film and particularly whether segregation takes place at the grain boundaries, it is not easy to obtain this microscopic information with the regular XRD and Raman measurements since no obvious new phase

formation is observed following the nitrogen implantation within the spectrometer's limitations. However the surface sensitive XPS and XANES characterizations may partially answer this question. For instance, we employed XPS to check the validity of nitrogen implantation into the VO_2 film during the sample preparation. After N_2^+ implantation of the VO_2 films, the samples experienced Ar^+ sputtering at 1 keV for 10 minutes and then checked with XPS measurements. N 1s XPS indicates a strong signal after the Ar^+ sputtering, suggesting nitrogen atoms are effectively implanted (see Figure R2). Moreover, the intensity of N 1s after sputtering appears comparable with that before the sputtering treatment. This suggests there is no strong segregation of the nitrogen atoms to the grain boundaries under our moderate experimental conditions.

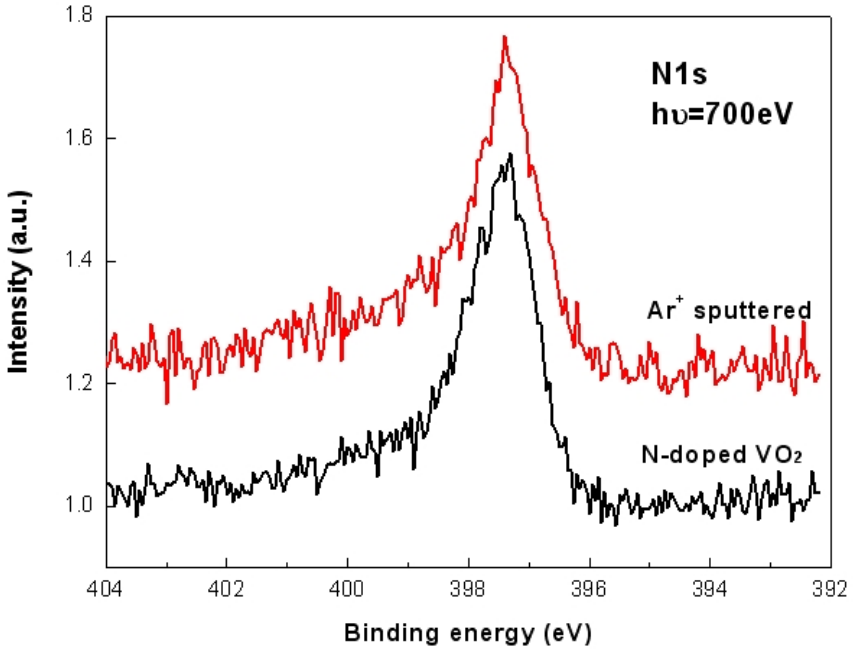

Figure R2 N 1s spectra excited with the photon energy of 700 eV. The upper curve is taken from the N-doped VO_2 after Ar+ sputtering at 1 keV for 10 minutes.