Growth control of metal-insulator transition and oxygen stoichiometry in VO_{2-x} epitaxial thin films

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Metal-insulator transition (MIT) in strongly correlated electron oxides (CEOs) is one of the most fascinating research topics in condensed matter physics, but is still far from fully understood.[1–3] Among CEOs, VO_2 has attracted great attention since it undergoes the MIT near room temperature (~340 K). Due to the multivalent nature of vanadium oxides,[4–12] *i.e.* $V_2^{+3}O_3$, $V^{+4}O_2$, and $V_2^{+5}O_5$, the oxidation state and oxygen stoichiometry play an important role for determining the structural and electronic properties. For example, Jeong *et al.* recently demonstrated suppression of the MIT in VO_2 by electric field-induced oxygen vacancies.[13] In addition, due to the lack of a good understanding on growth control of the oxidation state, stabilization of phase pure vanadium oxides with a single oxidation state is extremely challenging. Therefore, precise control of the chemical valence or oxidation state of vanadium in vanadium oxide thin films is highly desirable for not only fundamental research, but also technological applications

In this study, we precisely mapped out the optimal growth window of vanadium oxides with different oxidation states by the state-of-the-art pulsed laser epitaxy technique for providing a guideline to grow high quality thin films. As shown in Fig. 1, a well-pronounced MIT was only observed in VO_2 films grown in a very narrow range of oxygen partial pressure (10~25 mTorr). On the other hand, thin films grown either in lower (<10 mTorr) or higher (>25 mTorr) oxygen partial pressure resulted in the formation of V_2O_3 and V_2O_5 phases, respectively, as well as the suppression of the MIT for both cases. We have also found that the resistivity ratio before and after the MIT of VO_2 thin films can be further enhanced by one order of magnitude when the films are further oxidized by post-annealing at a well-controlled oxidizing ambient. This result indicates that stabilizing vanadium into a single valence state has to compromise with insufficient oxidation of an as grown thin film and, thereby, a subsequent oxidation is requisite for an improved MIT behavior.

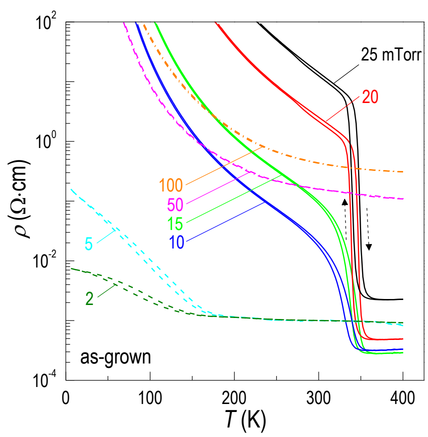

Figure 1 Resistivity $\rho(T)$ curves of as grown vanadium oxide films deposited at various oxygen partial pressure $P(O_2)$. Dashed, solid, and dash-dotted lines represent three vanadium oxide phases, *i.e.* V_2O_3, VO_2, and V_2O_5, respectively.

Our result ultimately stresses the importance of oxygen content in VO_2 that plays a critical role in the sharpness and resistivity ratio of the MIT. VO_2 thin films easily absorbed oxygen as low as 300 °C, implying that VO_2 can be regarded as an oxygen sponge.

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