Surface Stress-induced Domain Dynamics and Phase Transitions in Epitaxially Grown VO₂ Nanowires

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We demonstrate that surface stresses in epitaxially grown VO₂ nanowires (NWs) have a strong effect on the appearance and stability of intermediate insulating M₂ phases, as well as the spatial distribution of insulating and metallic domains during structural phase transitions. During the transition from an insulating M₁ phase to a metallic R phase, the coexistence of insulating M₁ and M₂ phases with the absence of a metallic R phase was observed at atmospheric pressure (See Fgiure 1). In addition, we show that for a VO₂ NW without the presence of an epitaxial interface, surface stresses dominantly lead to spatially inhomogeneous phase transitions between insulating and metallic phases. In contrast, for a VO₂ NW with the presence of an epitaxial interface, the strong epitaxial interface interaction leads to additional stresses resulting in uniformly alternating insulating and metallic domains along the NW length. In order to demonstrate the detailed structural changes and coexistence of two different insulating phases (M₁, M₂), we prepared naturally bent NWs with uniform local curvature and nonclamped (strain-free interface) on a c-cut sapphire substrate using a PDMS transfer method, which is a technique widely used for transferring graphene. Raman measurements were carried out at two featured positions; i) the non-clamped, straight part of a NW (A in the upper inset of Figure 4a), and ii) the largest bent part of a NW with the high tensile strain at the centre of the outer edge of the local curvature region (B in the lower inset of Figure 2a). As shown in Figure 2a, the evolution of Raman spectra obtained from the straight region of a NW (A), which exhibits direct structural changes from M₁ to M₂ phases, is quite similar to that measured on a transferred NW non-clamped shown in Figure 1. Interestingly, in the bent part of a NW (B), coexistence of both M1 and M2 phases as evidenced by peaks associated with only M₁ and M₂ phases were observed even at room temperature. As the temperature increases, the intensity of M₁ peaks gradually decreases and that of M₂ peaks continuously increases, showing the evolution of M₁-M₂ phases. When the temperature increases further, R phases start to appear.

References

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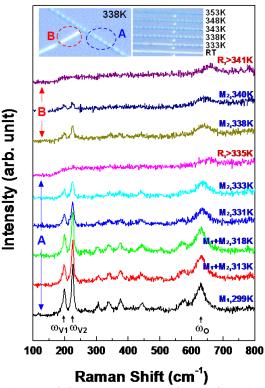


Figure 1. Temperature dependence of Raman spectra of transferred individual VO_2 NWs. As the temperature increases, phonon frequencies shift toward higher frequencies and phase transitions from an M_1 to an R phase occur spatially along the NW length. Raman spectra indicated by A, B and C were obtained from the circle region of the NWs, respectively (left inset). Optical images of bright and dark domain patterns corresponding to insulating and metallic phases, respectively (right inset), reveal spatial phase transitions along the NW length.

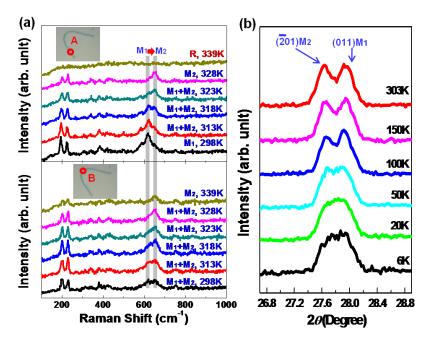


Figure 2. (a) Raman spectra obtained from the straight part (A in the upper inset) and bent part (B in the lower inset) of a VO_2 NW, which show the direct evolution of M_1 - M_2 phases. **(b)** Temperature dependence of XRD data from ensembles of epitaxially grown VO_2 NWs, measured during cooling from 303 K to 6 K. These results demonstrate that M_1 and M_2 phases can coexist with the absence of an R phase at atmospheric pressure.