



Surface-disorder and cationic-site defect-induced ferromagnetism and correlated Raman vibrational modes in $\text{Sn}_{0.9}\text{In}_{0.1}\text{O}_2$ nanocrystalline thin-films

Shyamsundar Ghosh

Department of Physics, Bejoy Narayan Mahavidyalaya, The University of Burdwan, P. O., Itachuna, Hooghly, 712 147, India

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ABSTRACT

Exploration of magnetism in otherwise nonmagnetic wide-band oxides through defect-engineering has been become a new challenge to achieve the goal of room-temperature ferromagnetic semiconductor for potential applications in spintronics. In this backdrop, we report surface-disorder and cationic-site defect-induced FM in series of $\text{Sn}_{0.9}\text{In}_{0.1}\text{O}_2$ nanocrystalline thin films prepared using pulsed laser deposition different Argon (Ar) pressure. With the increase of deposition pressure, surface morphology of $\text{Sn}_{0.9}\text{In}_{0.1}\text{O}_2$ nanocrystalline thin-films changes as nanoclusters, nanofibres, nanoflakes and nanowires respectively, crystallite size decreases and surface disorder increases gradually. Raman, X-ray photoelectron and Photoluminescence spectroscopic analysis confirm the formation tin-vacancy (V_{Sn}) and associated combinational defects that significantly increase the ferromagnetic signal in In-doped SnO_2 thin-films. The substitution of trivalent In^{3+} in Sn site also cause serious lattice distortion which transform the Raman inactive modes A_{2g} and E_u modes into Raman active modes and thus appeared at 574 cm^{-1} and 354 cm^{-1} respectively. The nanowires of $\text{Sn}_{0.9}\text{In}_{0.1}\text{O}_2$ are found to possess highest concentration of V_{Sn} defects and therefore it exhibits strongest ferromagnetic moment (M_s) $\sim 11.1\text{ emu/cm}^3$ and highest Curie temperature (T_C) $\sim 572\text{ K}$. The gradual decrease in carrier concentration with the increasing deposition pressure suggests stabilization of hole-mediated V_{Sn} -induced room temperature FM in series of In-doped SnO_2 nanocrystalline thin-films.

1. Introduction

Defect-induced ferromagnetism (FM) in low-dimensional (1D/2D) otherwise nonmagnetic wide-band semiconducting oxide nanostructures has been recently drawn prime research attention due to potential applications in spintronics, magneto-optical and magneto-electrical devices [1–3]. During growth of oxide-based 1D or 2D nanostructures or thin-films in diverse atmosphere, many structural defects like cation vacancies, oxygen vacancies (V_{O}), interstitials and antisite defects and dislocations can stabilize within lattice. Among them, some type of defects can induce local magnetic moment within host semiconductors [4,5]. In fact, there are significant reports on the evidence of room-temperature FM in ZnO and TiO_2 based-oxides associated either with transition-metal doping or oxygen vacancy defects [6–12]. On the other hand, many theoretical studies [13–16] have suggested that the formation of cation-vacancy defects rather than V_{O} defects in similar oxides can be more effective to stabilize carrier-mediated room-temperature FM. Density functional theory (DFT) by Elfimov et al. [13]

and Pemmaraju et al. [14], and later Osorio-Guillen et al. [15] has predicted that wide-band CaO thin-film can behave as pure ferromagnet with 5% Ca vacancies. Later, Zhang et al. [16] has shown experimentally that the Nb incorporation can help to stabilize Ti vacancy defects in pulsed-laser deposited Nb-doped TiO_2 thin film. Although they do not observed FM due to very low concentration of Ti vacancies and free carrier density. After that, Zn-vacancy induced FM is observed in different nonmagnetic element doped ZnO thin films [6,17], ZnO:Li nanorods [18] and also in our previous work on ZnO:K nanowires [9].

In this backdrop, tin dioxide (SnO_2) is an important transparent conducting oxide (TCO) which finds potential applications many field of science such as sensors, solar panel displays and other transparent electronic devices [19–21]. Unlike ZnO or TiO_2 , it has been less explored as far as defect-modified magnetic behaviours are concern. DFT calculations by Gul Rahman et al. [22] have explained tin-vacancy (V_{Sn}) with proper defect-engineering can generate large magnetic moment in SnO_2 . They have also suggested that observation of unexpected gaint magnetic moment (7.5 ± 0.5) μ_B/Co in Co-doped SnO_2 films by Ogale et al. [23],

E-mail addresses: sghoshphysics@gmail.com, shyamsundar@bnmv.ac.in.

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the work reported in this paper.

Data availability

Data will be made available on request.

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