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Influence of annealing temperature on the magnetic properties of Cr⁺ implanted AlN thin films



A. Shah^{a,*}, Arshad Mahmood^a, Zahid Ali^a, T. Ashraf^a, Ishaq Ahmed^b, Mazhar Mehmood^c, Rashad Rashid^a, Imran Shakir^{d,*}

^a National Institute of Lasers and Optronics (NILOP), PO Nilore, Islamabad, Pakistan

^b Experimental Physics Lab, National Center for Physics (NCP), Islamabad, Pakistan

^c DMME, Pakistan Institute of Engineering and Applied Science (PIEAS), PO Nilore, Islamabad Pakistan

^d Sustainable Energy Technologies (SET) center building No 3, Room 1c23, College of Engineering, King Saud University, PO-BOX 800, Riyadh 11421, Kingdom

of Saudi Arabia

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ABSTRACT

Diluted magnetic semiconductor (DMS) AlN:Cr films were produced by implanting various doses Cr⁺ ions into AlN thin films at room temperature followed by a thermal annealing process. The structural and magnetic characteristics of the samples were investigated as a function of annealing temperature by means of Rutherford backscattering and channeling spectrometry (RBS/C), X-ray diffraction (XRD), Raman spectroscopy, vibrating sample magnetometer (VSM) and SQUID. Structural analyzes demonstrate that implantation damages gradually decrease with the increasing of annealing temperature. Moreover, better recrystallization in the implanted part of the samples was observed for the sample annealed at 950 °C. Both XRD and Raman pattern illustrate that no secondary phase or metal related-peaks were appear in all the samples. Magnetic analysis reveals that annealed Cr^+ -implanted samples exhibit ferromagnetism at room temperature, however, the sample annealed at 950 °C shows improved magnetiz characteristics. The saturation magnetization is estimated to be 9.0×10^{-5} emu/g and the coercive field (H_c) is approximately 200 Oe for the samples annealed 950 °C. In SQUID analysis, FC/ZFC measurements indicate that the Curie temperature (T_c) is well above room temperature.

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1. Introduction

III-Nitrides based dilute magnetic semiconductors (DMSs), are thought to be ideal materials for spin-electronics or spintronics devices, since it utilizes both the charge and the spin of electrons simultaneously to create new functionalities beyond conventional semiconductor devices [1–3]. In particular, AlN has received intense attention in the emerging field of spintronics, due to its wide band gap and its predicted high Curie temperature (T_C) when doped with particular transition metals (TM). Moreover, AlN is an ideal host in this regard, since Kucheyev et al. [4] reported that AlN epilayer grown on sapphire substrates did not become amorphous even when implanted with heavy ions at high doses of keV energy. In addition, AlN ensures large sp–d hybridization between the valence orbitals and the d shells of the magnetic ions due to its small lattice constant. A theoretical study based on first principle calculation predicted that AlN-based DMSs could exhibit

E-mail addresses: attaullah77@yahoo.com (A. Shah), shakir@skku.edu (I. Shakir).

http://dx.doi.org/10.1016/j.jmmm.2014.12.032 0304-8853/© 2015 Elsevier B.V. All rights reserved. ferromagnetic behavior upon doping with transition elements [5–8]. In addition room temperature ferromagnetism has been experimentally observed in TM doped AlN DMSs [5,9–14].

The synthesis of ferromagnetic AlN could produce a wider range of all possible semiconductor spin-dependent devices. For instance, AlN:Mn acts as a ferromagnet which can be used as a magnetic barrier in a tunnel junction as well as a spin filter. The predicted Curie temperature of AlN:Mn is greater than 300 K and recently a Curie temperature of more than 340 K has been observed for AlN:Cr [14,15]. The Cr ions may be attractive for AlNbased DMS, since it has a lower vapor pressure than Mn, so it should have a larger sticking coefficient at elevated growth temperatures. Furthermore, the solubility of Cr in AlN is much higher than that of Mn, as the high solubility of transition metals in semiconductor matrices could facilitate modulating the magnetic properties of these materials.

The ferromagnetic Cr-doped AlN thin film have been prepared by several techniques such as magnetron sputtering [12,14], MBE [5,13,16] and ion implantation [17]. Among them, ion implantation provides a versatile and convenient method for introducing transition metals into semiconductors. Specifically it is widely used in

^{*} Corresponding authors. Fax: +92 51 2208051.

device fabrication steps such as selective-area doping, electrical isolation, etc. However, in all of these fabrication steps, limitations may arise due to implantation lattice disorder and its undesirable consequences [18].

Recently, the structural, optical and magnetic properties of Cr^+ -implanted AlN films have been studied by several groups [19–22]. However, there has been no systematic study on Cr ions implanted AlN thin films as a function of annealing temperature. In this study, we report on the structural and magnetic properties of Cr^+ -implanted AlN as a function of annealing temperatures.

2. Experimental details

The AlN thin epilayer was grown by RF-plasma-assisted molecular beam epitaxy (MBE) on sapphire (0001) substrate, as described in details elsewhere [19]. Implantation of Cr⁺ was carried out at energy of 750 keV at room temperature with doses of 5×10^{14} cm⁻², 1×10^{15} cm⁻² and 5×10^{15} cm⁻². The Chromium concentration is calculated around 5% as using program SRIM 2008 [23] and also confirmed by simulation of random spectra using RUMP [24]. Annealing of the implanted samples was carried out in order to recover implantation damages, re-crystallize the samples and activation of dopants. The samples were annealed at temperature 850 °C, 900 °C and 950 °C for 30 min in ambient N₂ gas.

Rutherford backscattering and channeling spectrometry (RBS/ C) was used to analyze the structure and crystal quality of the prepared samples. A 2.0 MeV He⁺ collimated beam produced by 5UDH-2 Pelletron were used for Rutherford backscattering and channeling. The detection angle was set to 165° and its resolution energy was about 18 keV. The angular channeling scans were carried out for two different crystallographic direction (0001) and $\langle -1102 \rangle$ in order to allocate the implanted atom in the host lattice.

The structural features of Cr⁺-implanted AlN films were further studied by means of XRD using the Bruker D8-Discover X-ray diffractometer with Cu k_{α} radiations of wavelength 1.5405 Å. The measurements were made in the 2θ angular range of 20–90°, with a step size of 0.02° and a scan rate of 2°/min. Raman measurements of the samples were performed at room temperature using He–Cd laser ($\lambda \sim 442 \text{ nm}/80.0 \text{ mW}$) as an excitation source. The signals collection and detection was carried out in the scan mode by the same objective lens using an air cooled $(-50 \circ C)$ CCD detector. The source peak (442 nm) was eliminated by using 442 nm cutoff filter. The spectrum was taken within the range from $500\ \text{cm}^{-1}$ to 950 cm^{-1} using DM320 monochromator and ANDOR DV 401A BV CCD software. The magnetic measurements were carried out at room temperature using Princeton Measurement Corporation vibrating sample magnetometer (PMC, VSM). In all of the magnetization measurements, the magnetic field was applied parallel to the sample plane. The diamagnetic contributions from the Al₂O₃ substrate to the total magnetization were subtracted.

3. Results and discussion

3.1. RBS/C spectrometry

Fig. 1 represents the RBS/C spectra for the as grown AlN and Cr^+ -implanted AlN films at various doses (5×10^{14} , 1×10^{15} and 5×10^{15} cm⁻²). Clearly a broad damage peak (centered at 0.95 MeV) beneath the film surface can be seen in the implanted samples. This broad peak (damage) is deduced to be the consequence of He⁺ being backscattered from the displaced Al atoms in the damage region. Moreover, the intensity of this damage peak increases with the increasing of ions fluence, which means that the density of displaced atoms increases with the increasing of

Fig. 1. RBS/C spectra of the as grown AlN film and $\rm Cr^{+-}implanted$ AlN films at various doses.

implanted ions dose. Similarly the incorporation of Cr atoms into the AlN matrix is also confirmed from this graph as the peaks corresponding to the Cr atoms appear in the spectra.

The channeling spectra of the Cr⁺-implanted AlN film with dose 5×10^{15} cm⁻² and subsequently annealed samples at 850 °C, 900 °C and 950 °C are shown in Fig. 2. The figure clearly demonstrates that the intensity of the broad damage peak decreases with the increasing of annealing temperature indicating a reduction in implantation related defects. These damages cannot be recovered completely at higher annealing temperature. However, in all cases 950 °C is the best annealing temperature for the possible recovery of the damages.

The channeling angular scans of the Cr⁺-implanted AlN sample with dose 5×10^{15} cm⁻² before and after annealing at 950 °C, recorded in (0001) and (-1102) crystalline directions are shown in Fig. 3. It shows that Cr atom has similar profile as that of host Al atom, which indicates that most of the Cr atoms occupy Al sites with slightly displaced from their equilibrium position. However, the narrow angular width for Cr points out the existence of implantation damages as already observed in the RBC/C spectra. The widths for the Cr-scan increase after annealing showing the reduction in implantation defects, whereas the width of Al-scan remains unchanged after annealing.



Fig. 2. RBS/C of Cr⁺-implanted AlN at $5 \times 10^{15} \, cm^{-2}$ and annealed samples at different temperatures.



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